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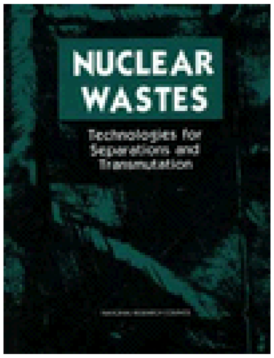

Nuclear Wastes: Technologies for Separations and Transmutation

Committee on Separations Technology and Transmutation Systems, National Research Council

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THE NATIONAL ACADEMIES
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Nuclear Wastes

Technologies for Separations and Transmutation

Committee on Separations Technology and Transmutation Systems
Board on Radioactive Waste Management
Commission on Geosciences, Environment, and Resources
National Research Council

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This report has been reviewed by a group other than the authors according to procedures approved by the Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

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Preface

Beginning in the 1940s, the U.S. government engaged in a very vigorous program of research, development, and utilization of nuclear energy. One part of the program was focused on the design and production of nuclear weapons. The other part of the effort was focused on the development of nuclear power reactors for both military and civilian use. From the beginning it was clear that use of nuclear energy would result in the generation of considerable amounts of high-level waste, containing radionuclides that are intensely radioactive and/or have long half-lives. Though it was recognized quite early that high-level waste would have to be managed safely without unacceptable risks to humans, for many decades other parts of the nuclear program (including interim storage) were given higher priority. The spent reactor fuel was stored in pools at the reactor site, and the high-level liquid wastes resulting from processing of defense reactor waste were put into underground tanks as temporary solutions to the problem. The age of the oldest of these "temporary" tanks containing processed liquid wastes is now approaching 50 years. Spent civilian-reactor fuel storage has been uneventful, but the high-level waste tanks are corroding and some have developed leaks, creating a hazardous situation. At many reactor sites these storage pools are nearly full—a situation requiring the construction of additional on-site pools or dry storage facilities.

It is expected that by about 2010 the U.S. civilian nuclear power reactors will have produced about 63,000 Mg (about 70,000 tons) of spent fuel. This will contain about 90% of all high-level radioactive waste in the U.S. nuclear program. The remaining spent fuel will result from the military program.

Extensive work has been done on research, development, and evaluation of methods for the ultimate safe disposal of this spent fuel and high-level radioactive waste. These studies have almost all concluded that the most practical approach would be to vitrify the liquid high-level wastes in a glass matrix. The resulting glass would then be encapsulated in suitable containers and buried in a specially selected and evaluated stable geological formation deep underground. Present U.S. policy is that spent power reactor fuel would also be encapsulated and buried.

To implement the disposal process, several recent administrations have announced their intention to establish a national repository for high-level radioactive waste. They developed a program schedule and passed legislation to provide the funding mechanism needed to carry out the program. However, the schedule for the program has been marked by continuous delays, and successive plans have had to be abandoned, resulting in billions of dollars of wasted expenditure. Some reasons for this dilemma are technical, but many are nontechnical. The current stalemate on the Yucca Mountain site in Nevada, sponsored by the U.S. Department of Energy (DOE) is an example of such a case. When Yucca Mountain was identified by the U.S. Congress as the first site to be evaluated, the Nevada governor initiated legal steps to try to stop the federal government from taking any steps to evaluate the site, even though it is on federal property.

Due to legal interventions such as this, the DOE-sponsored work to establish Yucca Mountain as the first U.S. high-level waste repository is over budget and behind schedule. Still in its investigative phases after many years of effort, it has not yet even been determined if the site is suitable.

Because of all these difficulties, DOE continues to explore other disposal options that might circumvent such problems. Some of the proposed solutions have focused on separating the hazardous long-lived radioactive nuclides in the waste and transmuting them by neutron bombardment to form nuclides that would be either stable or radioactive with a much shorter half-life. During the last decade, there has been a renewed interest by a few countries in such proposals, and some technological progress has been reported. This has led several of the DOE national laboratories to reexamine this concept to see if it might be put to practical use. Although this approach is technically feasible, its use involves several practical problems—a major technical problem is the requirement for production of very high neutron source intensities, which poses severe engineering and material challenges. Another difficult problem is achieving the very high efficiency required in the chemical separation of the radionuclides to be transmuted from the other wastes. Unless high separation efficiency is obtained, full benefits of transmutation may not be achieved. The residual waste may still be contaminated to the extent that it must be considered as high-level waste. In addition to these technical problems, the issues of economics, licensing, and public acceptance present significant obstacles to the adoption of transmutation systems.

As part of the reexamination of separations and transmutation (S&T) technology systems, in 1991 the Secretary of Energy, Admiral James D. Watkins, Jr., requested the National Research Council (NRC) to conduct a broad systems review of the application of S&T concepts to radioactive waste disposal. To implement this request, the NRC formed a 19-member multidisciplinary committee on Separations Technology and Transmutation Systems (STATS), under the direction of the Board on Radioactive Waste Management. The committee was assisted by a subcommittee on Separations, under the NRC's Board on Chemical Sciences and Technology, and a subcommittee on Transmutation, under the NRC's Board on Energy and Environmental Systems. These subcommittees included 10 additional experts. A third STATS subcommittee on Integration functioned until May 1992.

The expertise of these committees included engineering, physical and chemical sciences, accelerator physics, radioactive and hazardous waste disposal technologies, health and safety assessment, economics, public and regulatory policies and procedures, and plant operations.

The scope of the study was to prepare a reviewed report evaluating the relative effects, costs, and feasibility of employing separations and transmutation technologies in the Department of Energy's programs for managing (1) spent nuclear fuel from civilian power reactors, and (2) radioactive wastes in tanks at selected existing defense production reactor sites.

To initiate the study, an international workshop was held in January 1992, where the committee was briefed on the current status of knowledge in the field by experts representing a wide cross section of disciplines. Following the workshop, the committee undertook an in-depth evaluation of all the aspects concerned with separations technology and transmutation through a number of meetings and briefings. It met with representatives of laboratories and organizations involved with the work and visited their sites in the United States and abroad.

In May 1992, in response to a request by the Secretary of Energy, an interim report of the committee was submitted that summarized the committee's information at that time concerning actinide burning in advanced liquid-metal reactors and the possible impacts on the repository program.

The committee examined the currently used "once-through" fuel cycle versus proposed separations technology and transmutation systems for dealing with high-level radioactive waste. Two principal transmutation concepts involving critical reactors, both light-water reactors and advanced fast reactors were examined. In addition, a system was reviewed in which an accelerator combined with a subcritical reactor assembly produced neutrons. The committee also examined in detail the applicability of S&T in the management of defense wastes stored in tanks at the Hanford site.

This volume constitutes the final report of the STATS Committee. It provides a comprehensive evaluation of separations technology and transmutation technologies, its current status, and some insight on future potential. Associated topics dealing with health, safety, proliferation, socioeconomical, and regulatory issues are covered, as are possible schedules and economics of potential S&T systems. The report further examines whether implementation of S&T concepts can contribute to a safer and more economical waste disposal option than the present "once-through" fuel cycle and waste disposal option.

The report begins with an Executive Summary followed by a brief introduction to S&T technology in Chapter 1. Chapter 2 gives a description of the technologies involved. Chapters 3 and 4 describe the current status of separations and transmutation technologies, respectively. Some of the issues associated with defense waste are covered in Chapter 5. A number of important issues including licensing, health and safety, proliferation, and economics are reviewed in Chapter 6. Those desiring a more technical discussion will want to refer to Appendices C through K and Appendix P. Because of the frequent use of acronyms, the reader may find Appendix O valuable.

The STATS report had the benefit of the time and talents of highly professional and dedicated committee members, and it was my privilege and pleasure to work with them on this project. I acknowledge with thanks the many long hours and arduous work they provided in researching, writing, and revising their contributions to the many complex issues involved in separations technology and transmutation systems. Similarly, I extend my appreciation to the Department of Energy, which funded the study and express my appreciation to its staff for their technical support for the committee's work. On behalf of the committee I also express gratitude to the many scientists and engineers representing various organizations and laboratories who briefed the committee and supplied it with the required technical reports dealing with research, development, and evaluation studies on the subject.

The Subcommittee on Transmutation was chaired by Ed Kintner. The Subcommittee on Separations was chaired by Fred McLafferty. Edward Mason was chairman of the Subcommittee on Integration. My special thanks go to the chairs and members of the subcommittees for their valuable contributions to the STATS committee work.

John M. Googin, a STATS Committee member, died in January 1994. His vast knowledge in engineering and chemical sciences and extensive corporate memory made his contributions to the STATS Committee invaluable. We grieve his loss.

The successful conclusion of a study of this magnitude is very dependent on the skills, dedication, and energy of the NRC staff who were assigned to the committee. In particular I thank K.T. Thomas, STATS Study Director and Senior Staff Officer of the Board on Radioactive Waste Management for his many valuable contributions to the study. His pleasant personality, dependability, and talents were responsible for many improvements in the final report. Carl A. Anderson, Director of the Board on Radioactive Waste Management, also made many contributions, particularly to the interim report.

Thanks also go to James J. Zucchetto, Staff Officer for the Subcommittee on Transmutation, and Scott T. Weidman, Staff Officer for the Subcommittee on Separations, and their respective directors of the two boards, Mahadevan Mani and Douglas J. Raber. Finally, I thank Lisa Clendening, Verna Bowen, and Patricia Jones of the Board on Radioactive Waste Management for their valuable work and guidance in seeing this report through to completion.

NORMAN C. RASMUSSEN, *Chair*

Committee on Separations Technology and Transmutation Systems

Contents

APPENDIXES

Nuclear Wastes

Executive Summary

For over three decades, nuclear technologists have sought to develop practical methods for the safe management and disposal of highly radioactive wastes containing long-lived radionuclides. Some of the proposed solutions have focused on separating the hazardous long-lived radionuclide components of the waste and transmuting them by neutron bombardment to form nuclides that would be either stable or radioactive with a much shorter half-life. The Department of Energy's (DOE) financial support has been given in the past and/or sought for in the future by several proponents with proposals to develop separations technology and transmutation (S&T) systems. The Committee on Separations Technology and Transmutation Systems (STATS) of the National Research Council was formed at the request of DOE to evaluate the state of the art of S&T concepts. The purpose is to determine if S&T alternates offer advantages over the current government policy for the disposal of commercial spent fuel arising from the "once-through" light-water reactor (LWR) fuel cycle. The committee, in further fulfillment of its mandate, also reviewed processing options for the safe management of high-level waste (HLW) generated by the defense programs, in particular, addressing the special problems in dealing with such wastes at the DOE facility in Hanford, Washington.

THE "ONCE-THROUGH" FUEL CYCLE

The present national plan is to store and "decay cool"[1] the spent fuel at reactor sites for a decade or more and then to ship it to a monitored retrievable storage (MRS) facility where it would be temporarily stored and eventually encapsulated and shipped to a geological repository. It is called the "once-through" fuel cycle because the spent fuel is not reprocessed and thus not recycled. All the other fuel cycles involving S&T systems reviewed in this report require reprocessing with its associated implications.

An advantageous feature of the direct disposal of the spent reactor fuel in a repository without reprocessing is that if reducing conditions exist in the repository, the actinides are not readily moved in groundwater pathways; they are quite insoluble under such conditions. However, for some invasive scenarios, e.g., human intrusion or under oxidizing conditions in the repository, actinide release may occur from the repository and cause some risk. Otherwise, the principal doses to humans after long periods of time are due mainly to the fission products technetium (^{99}Tc) and iodine (^{129}I) that are water soluble and so are moved through groundwater pathways.

Currently, DOE is examining a site at Yucca Mountain, Nevada, to determine if it can be licensed as the first HLW repository. The U.S. Nuclear Regulatory Commission (NRC) regulations for a licensed repository require that the encapsulation of the spent fuel remain effective for at least 300 to 1,000 years, which is a subordinate requirement to the longer limits on release rates—typically 10,000 years or more. By statute,[2] the first repository is limited to a loading of 70,000 Megagrams of heavy metal (MgHM) until a second repository begins operation. Current designs call for 62,000 MgHM of the total capacity to be allocated for the commercial LWR spent fuel expected to be produced by the year 2010. This capacity will provide for approximately 60% of the spent fuel expected to be discharged from the

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[1] During the storage period, some of the short lived heat emanating radionuclides will decay, thus reducing considerably the heat output from the spent fuel.

[2] The U.S. policy governing the disposition of spent fuel from commercial reactors is provided by the Nuclear Waste Policy Act of 1982 and the Nuclear Waste Policy Amendments Act of 1987.

existing LWRs. The rest of the capacity would be devoted to vitrified defense HLW. This capacity is not expected to be sufficient for the disposal of the total amounts of defense wastes produced. By coincidence, it happens that the 62,000 MgHM happens to be the amount to be produced in the spent fuel of the LWRs by the year 2010.

THE S&T CONCEPTS

The committee has reviewed the three principal transmutation concepts for commercial reactor spent fuel for which considerable information is available. These concepts use a light-water reactor (LWR), an advanced liquidmetal reactor (ALMR), or an accelerator-driven subcritical nuclear reactor concept for producing neutrons called the accelerator transmutation of waste (ATW). Two other proposals, the particle bed reactor (PBR) and the Phoenix accelerator-driven reactor concept, did not have sufficient detail for an adequate comparison with the principal three. The sixth proposal, the Clean Use of Reactor Energy (CURE), is a study of S&T with fast reactors and LWRs, and does not rely on one specific transmutation scheme. The PBR, Phoenix, and CURE concepts are discussed in the appendices but do not appear in any detail in the main report. However, the issues raised by the PBR and Phoenix proposals and the CURE study are covered in the evaluation of the principal three concepts.

In comparing the principal concepts, the committee assumes that the mission of each is to provide for preparation and transmutation of about 600 Mg of transuranics (TRUs) contained in the 62,000 MgHM of existing LWR spent fuel, as well as packaging and storage of selected fission products contained in the accumulated spent LWR fuel that is otherwise destined for the first geologic repository. The LWR and the ALMR concepts are also evaluated for their abilities to transmute only the plutonium fraction of the TRUs. The prospective benefits and deficiencies for waste disposal of such evaluations are examined for the full S&T and for the once-through LWR fuel cycle.

To develop some figures of merit for comparison among the different S&T concepts, the committee looked at two different scenarios relative to the present once-through LWR fuel cycle. The first, called the declining nuclear power or "phase-out" scenario assumes that the TRUs and selected fission products are transmuted and that all nuclear facilities are shut down as quickly as practicable, consistent with S&T requirements. The second, called the continuing nuclear power scenario, assumes that the amount of nuclear power in the United States stays constant, i.e., at about its present level of 100 GWe. In that scenario, some fraction of the reactors would be operated to transmute the TRUs in the accumulated LWR spent fuel, but with some continuing TRU production. These alternate scenarios are analyzed to indicate the implications of transmutation on the power profiles and TRU inventories in the fuel cycle over time and the implications for waste disposal.

OVERALL CONCLUSIONS

The committee found no evidence that applications of advanced S&T have sufficient benefit for the U.S. HLW program to delay the development of the first permanent repository for commercial spent fuel. The committee believes that the thermal neutron flux of a LWR and the fast flux of an ALMR could be used to transmute the TRU isotopes in spent reactor fuel. If the proposed ATW could operate with the currently proposed characteristics, its thermal flux would also be effective in the transmutation of the TRUs. The fission products ^{99}Tc and ^{129}I could also be transmuted by either the LWR or the ATW. The high thermal flux of the ATW would be an advantage in this process. Although a significant fraction (90 to 99%) of many of the most troublesome isotopes could be transmuted, this reduction of key isotopes is not complete enough to eliminate all the process streams containing HLW, so the need for a HLW repository is not eliminated. However, the total HLW storage capacity required would be reduced. Transmutation, thus, would have little effect on the need for the first repository.

In view of the above, the committee concluded that the once-through LWR fuel cycle should not be abandoned. Further, this has the advantage of preserving the option to retrieve energy resources from the wastes for an extended period of time. This can be achieved by adopting a strategy that will not eliminate access to the nuclear fuel component of the waste for a reasonable period of time, say about 100 years, or by preserving easy access to the repository for a prescribed period of time, or by extending the operating period of the repository.

A reason for supporting continued use of the once-through fuel cycle is that it is more economical under current conditions. Some analysts predict that future demand for uranium—and as a consequence its price—may increase to a point where recycling becomes economically competitive. Should this happen, the choice of once-through fuel cycle would have to be reexamined (see Appendix F).

The committee concludes that over the next decade the United States should undertake a sustained but modest and carefully focused research and development program on selected S&T technologies, with emphasis on improved separations processes for separating LWR and transmuter fuels beyond the existing plutonium and uranium extraction (PUREX) process and for fuels containing more actinide elements and selected fission products. These conclusions apply for either the continuing or phase out modes of the S&T systems.

DETAILED FINDINGS AND CONCLUSIONS

Findings Regarding Potential Radiological Effects of S&T

Thermal neutrons produced by an LWR or ATW could reduce the amount of ^{99}Tc and ^{129}I and thus reduce long-term doses from repository water pathway releases. The fast neutrons from the ALMR do not effectively transmute these two long-lived fission products. However, modifications of core design involving specially moderated fuel assemblies could reduce the fission product inventories of these two fission products.

The transmutation of actinides using an LWR, ALMR, or ATW could reduce the doses from repository water pathway releases. To the extent that reducing conditions dominate within or very close to the waste package, the actinides are expected to be insoluble and the doses so small that transmutation would have little effect. If oxidizing conditions were to dominate within a waste container and externally, neptunium might be sufficiently soluble so that transmutation could reduce neptunium doses substantially (see chapter 2 and Appendix G for details).

The transmutation of actinides using an LWR, ALMR, or ATW could reduce the repository doses resulting from certain invasive scenarios, e.g., volcanoes, human intrusion, etc. Many of these scenarios can also be dealt with to some extent by the choice of disposal site and repository designs. It must be recognized that this approach requires the balancing of many attributes.

To the extent that actinides are used as reactor fuel, the amount of uranium that needs to be mined, and therefore short-term radiation exposures from mining and milling, will be reduced. However, there will be a short-term increase in radiation exposure from reprocessing and other fuel-cycle activities not part of the once-through cycle. The population doses from each of these sources are thought to be small compared to natural background doses.

If the radioisotopes of concern were separated from the spent fuel and incorporated into a waste form better than spent fuel, reductions in long-term dose may be possible. This would involve the chemical processing needed to prepare insoluble compounds of the radionuclides of interest and integrating them into glass or some other matrix. All of this may be within the capability of modern chemical technology, but remains to be demonstrated. Qualification of a new waste form would require a significant research and development effort.

Conclusion Estimates of changes in dose from any of the above actions are small. The National Council on Radiation Protection and Measurements estimates 0.3 latent cancer fatalities/GWe-yr from the whole LWR fuel cycle. The estimate of the Oak Ridge National Laboratory for the dose reduction is 0.06 cancer fatalities/GWe-yr from the whole LWR fuel cycle. These calculations include a dose commitment integrated 1,000 years into the future from milling operations (see Chapter 6). Taken alone, none of the dose reductions seem large enough to warrant the expense and additional operational risk of transmutation.

Findings Regarding Separations Technology

Separations Technology Requirements for ALMR and LWR

For aqueous separations a combination of PUREX solvent extraction processes for actinides up to plutonium followed by a TRU extraction process for actinides above plutonium (TRUEX) could be used. Further research and development would be required before an economic commercial-scale TRUEX-like plant for S&T applications could be built.

If the proposed S&T systems' goal of eliminating HLW were to be achieved, the overall process losses would have to be reduced below those of current commercial systems.

ALMR/IFR proposes to develop pyroprocessing of ALMR and LWR spent fuel to reduce cost. The process has been demonstrated on a few kilogram scale for metallic fuel. Work has been done on gram quantities of simulated UO_2 fuel. Work on pyroprocessing of kilogram amounts of UO_2 continues in order to determine if the desired savings can be achieved.

Some remaining problems of pyroprocessing of UO_2 are conversion of oxide to metal, separation of cladding, and processing low concentrations of TRU.

Promising work is under way on improved separations systems. As yet none has been tested at the pilot-plant scale (details in Appendix D).

Separations Technology Requirements for the ATW

The goal of ATW is to transmute the inventory of radioactivity so all TRU waste leaving the site could be treated as LLW. The program calculates that process losses of 0.02% for plutonium and neptunium, and 0.0003% for americium and curium, must be achieved for both the LWR spent fuel and the on-line reprocessing system to be successful. To date, such levels have not been achieved on a commercial scale.

The aqueous ATW has very short cooling times so the separations must take place in very high radiation fields that would cause serious degradation of both water and organic agents. Processing using molten metals or salts avoids the problems of radiation degradation because these systems are very resistant to radiation damage.

Other severe problems are shared by both the aqueous and molten-salt processing proposals. They include severe decay heating from some of the separated actinides and nonuniform phase and thermal mixing in both slurry and molten salt.

Conclusion A number of the LWR and ALMR goals that depend on separations technology have yet to be fully demonstrated. Of particular importance is a reduction in the overall process losses during separations. Thus, research and development in separations technology must be an essential part of any program to develop S&T.

For ATW, processes based on molten-salt and molten-metal systems are very radiation-resistant and would not be subject to radiation-induced reagent degradation as is the case for aqueous systems. The ATW separations concepts that have been proposed are beyond demonstrated technology and are questionable in the intermediate term.

It is improbable that the ATW system would be able to achieve the goal of no long-lived HLW output because the unavoidable process losses would lead to contamination of various parts of the system. Eventually, these items would have to be treated as HLW or treated by additional expensive processes.

Findings Regarding Transmutation Technology

Current Status of LWR, ALMR, and ATW

The LWR has several thousand reactor years of experience as a power generator. If fission product transmutation were to be done using LWRs, modifications would be mainly in the fuel cycle and blanket. There would be a high level of confidence that the system would meet design objectives.

The ALMR would be based on breeder reactor technology that has been under development for over three decades. Design and prototype experience are sufficient to allow a high degree of confidence that a successful transmuter could be designed and built. Considerable research and development would be required (1) to assure that the low breeding ratio of a burner would not create safety problems and (2) to assure that the changes in fuel composition caused by burn-up would not adversely affect performance and safety.

The ATW system offers a higher burn-up rate but has a design that departs from previous experience by wide margins in a number of areas. These include on-line reprocessing of fluid targets, fluences of energetic particles yielding 100 displacements per atom, and very high thermal neutron fluxes: greater than 10^{15} neutrons/cm^2s. The designers of the ATW propose either slurry or molten-salt systems. Both types have had serious materials problems at the end of their abbreviated development programs. The target and the subcritical assemblies must dissipate as much as 8,000 MW of thermal power, raising difficult cooling problems.

Conclusion The ATW is in a far less developed state than the LWR or the ALMR. To prove the viability of the concept would require extensive research and development. Some of the design criteria are at the limits of what has been achieved. The system includes a large accelerator to produce neutrons, an 8,000-MW subcritical reactor, an on-line reprocessing system, and the operation of a 1,600-MWe power plant. To achieve reasonable on-line availability of such a system would be a major challenge.

Relative Capabilities of the Concepts for TRU Transmutation

Since TRUs can be transmuted in either a fast or thermal neutron spectrum, any of the three concepts could be used to transmute TRUs. The neutron source of choice will depend upon the composition of the TRUs and the desired end point of the irradiation. Some of the important considerations are discussed below. It should be noted that the transmutation of long lived fission products (e.g., ^{99}Tc and ^{129}I) is most readily accomplished by thermal neutrons from the LWR and ATW concepts. It would be possible, however, to have a neutron moderating target region in a ALMR to transmute these fission products.

ALMR

The ALMR transmuter has been proposed to reduce the amount of TRUs in waste below the limiting amount that could be released to the environment according to Environmental Protection Agency (EPA) standard 40CFR191—until recently the assumed standard for a Yucca Mountain repository.[3] For ^{239}Pu, the most abundant radionuclide to be transmuted, the corresponding amount proposed in the waste would be less than 0.03% of that in LWR spent fuel. The ALMR/IFR project has not described how such low losses could be attained in any of its published reports.

For the ALMR, the limiting breeding ratio is 0.65; below this, there may be safety problems because of a positive void reactivity coefficient. For an ALMR with a 0.65 breeding ratio and fueled with TRUs from LWRs for the constant power scenario, it would take about two centuries of operating time to reduce the inventory of the residual TRUs to about 1% of the inventory in the reference LWR once-

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[3] Although the EPA 40CFR191 standard no longer applies to the proposed Yucca Mountain repository, it is possible that such release limits, or the equivalent thereof, could still become part of the new standard. Also, the ALMR/IFR project makes the assumption that EPA release limits applicable to LWR spent fuel could be used as such for HLW generated in transmutation.

through fuel cycle of the same electrical power. The first century of constant-power operation could reduce the TRU inventory fraction to about 14% of the original. In a declining power scenario, the TRU inventory fraction could be reduced to as low as 9% in 100 years.[4]

LWR

To achieve the EPA standards of 40CFR191 for a constant power scenario, the LWR transmuter would require slightly less time than would the ALMR of 0.65 breeding ratio. However, the lower per-cycle processing loss required for americium and curium would be less than is achievable with present technology. In the declining nuclear power scenario the LWR transmuter could similarly reduce the actinide inventory fraction to about 9% in 100 years.

The length of time to reduce TRU inventories in a plutonium-burning LWR relative to the once-through LWR fuel cycle would be shorter than that for the nonbreeding ALMR.

ATW

The ATW depends on very low per-cycle process losses to achieve a high TRU inventory reduction. However, the frequent processing of the fluid fuel will result in the build-up of plutonium and neptunium in the ATW waste to as much as 0.4% of that in the spent-fuel waste of the reference LWR once-through fuel cycle of the same electrical power production capacity.

It is estimated to take 1,400 years and 90 years, respectively, for the constant and declining power (phase-out) scenarios to reduce the TRU inventories in the aqueous ATW reactor to the levels required by EPA standards. For the nonaqueous ATW the corresponding times would be 350 years at constant power and about 50 years with declining power.

The inventories of residual plutonium, neptunium, americium, and curium in the ATW reactor systems would require disposal in a geologic repository, as would other ATW waste such as Zircaloy pressure tubes from the aqueous ATW.

It would be impossible for ATWs to effectively eliminate all long-lived radionuclides to a degree that important residual radioactivity would persist no longer than a human lifetime as has been asserted by the ATW proponents.

Conclusion The proposed S&T systems require many decades to centuries to achieve a significant net reduction in the total TRU inventory relative to that of a once-through LWR fuel cycle. Comparing S&T systems using LWRs, ALMRs, and ATWs of the same electrical power production capacity, the ATW projects the highest TRU consumption rates. TRU consumption rates for an ALMR increase significantly as its breeding ratio is lowered to 0.65 from the conventional value near unity. The LWR—specially designed for burning plutonium— would have a net TRU consumption rate similar to an ALMR of 0.65 breeding ratio. As an alternative to S&T of all TRUs, reprocessing the LWR spent fuel and recycling only the plutonium to LWRs or ALMRs—using improved low-loss waste forms for the other HLW constituents—would be simpler and perhaps somewhat less expensive to implement.

Findings Regarding Feasibility, Development, and Deployment

LWR

Current LWR designs could be modified for transmutation of most actinides and the more troublesome fission products. This system has the shortest time and lowest cost to complete development and full-scale demonstration of technical performance and system costs—about 8 to 10 years at a level of effort very roughly estimated at perhaps $50-$100 million per year, exclusive of the cost of construction of any major test facilities.

ALMR

The ALMR, under development for decades as a fast breeder reactor, could be modified to operate as a TRU transmuter. Transmutation of fission products is not practical with an ALMR. An ALMR/IFR with integral pyroprocessing of its spent fuel has not yet been demonstrated at pilot-scale, but it may be more economical than an aqueous system. Considerable experimental and calculational effort at breeding ratios of less than unity (required for actinide burning) would be necessary to develop the proposed ALMR/IFR concept.

Compared to an LWR, an ALMR/IFR has a somewhat higher cost to complete development and demonstration—about 15 to 20 years at a level of effort very roughly estimated at twice the cost of LWR per year, exclusive of the cost of construction of any major test facilities. This falls between the LWR and ATW estimates.

ATW

The ATW poses major engineering and materials challenges due to the extraordinary operating conditions in the reactor and beam target. Heat removal is a major issue for development as well as for safety and licensing. Also, the possibility of reactor transients, unevaluated in any detail at

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[4] The "inventory reduction fraction" is the ratio of the TRU inventory in the transmuter, fuel cycle, and waste to that in the spent fuel of a reference LWR once-through fuel cycle of the same electrical production.

present, may require means of reactivity control beyond that afforded by merely turning off the accelerator. The ATW concepts use fluid fuels, which involve unproved technologies for fuel fabrication and on-site reprocessing and which raise a number of operational and safety issues for which detailed analyses have not yet been done. Questions have been raised about whether it would be simpler to make the subcritical assembly a critical assembly, thus eliminating the need for the accelerator and beam target and resulting in simpler core designs and more economical systems (see Chapter 4). Extensive research and development would be required even to ascertain whether an ATW is technically feasible. If found to be so, the ATW may require more than 20 years of development at an uncertain but relatively higher cost for development and full-scale demonstration.

Time Scale for Deployment. Assuming that development can be completed and that favorable institutional arrangements can be achieved, the licensing, construction, and initial operation of an S&T system based on LWRs or ALMRs of sufficient scale to *begin* to affect spent-fuel emplacement in a geologic repository would require one to two decades after a demonstration of system feasibility.

To permanently achieve the benefits of S&T to the repository and nuclear fuel cycle, the system would have to operate for many decades. If the system was not producing the desired results and it was decided to stop transmutation, it may, in principle, be possible to convert the LWRs and ALMRs in service back to the most optimum fuel cycle. In this case, some of the investment in reactors may not be lost. However, the recycling equipment, including the reprocessing plants, fuel fabrication facilities, and other equipment might be a loss. In addition, it is doubtful that ATWs would be used for electricity production in view of the very low thermal efficiency attainable.

Conclusion The S&T systems differ widely in their state of technological maturity and present a broad spectrum of development issues, risks, costs, and schedules. The most mature system concept for transmuting TRUs, that which uses LWRs, needs fuel-cycle development and would require about a decade and significant financial resources to reach the point of deployment. Compared to the LWR system, an ALMR/IFR system for transmuting TRUs would require more financial resources and take longer, perhaps a decade and a half, to reach the point of deployment. The ATW concepts require major development before the chances of success can be realistically assessed. An S&T system of appropriate scale must be operated for many decades to achieve the permanent benefits to the repository and other parts of the nuclear fuel cycle. If S&T operations were shut down early, facilities related to recycling (i.e., reprocessing and refabrication plants) are likely to be a loss.

Findings Regarding System Integration

The Nature of an S&T System

An S&T system capable of having a significant effect on the waste disposal problem would require a number of large nuclear facilities. The facilities needed include 10 to 20 transmuting nuclear reactors that would substitute for a like number of once-through LWRs, so that the total amount of electricity production would be the same. In addition, one or two reprocessing plants, remotely operated fuel fabrication capacity, and waste-packaging facilities would be required. Mechanisms for transportation between the facilities would have to be developed and implemented. Since the feedstocks of one part of the system are dependent on the output of the other parts, efficient operation will depend on integrated operation of the various parts. Those feedstocks that are in transit or storage are not being irradiated, and therefore, the overall transmutation rate is lowered. The usual practice of maintaining a stockpile of feedstock between the stages in the process will thus incur a penalty in transmutation rate. For this reason, integrated operation of the system to maximize the plant transmutation rate will be a difficult problem.

Conclusion An S&T system would require integrated operation of a number of facilities: about 80 LWRs, 20 transmuters, 2 reprocessing plants, and a few refabrication plants as compared to about 100 LWRs in the once-through system. The integrated operation of the system for the optimum transmutation rate will present an additional difficult problem for the S&T cycle.

Findings Regarding Economics

Issues

Implementation of an S&T system for nuclear waste burning would cost more for the same electrical production capacity than the corresponding system using the same type of reactor but optimized for power production. This occurs because (1) reprocessing would be required for a transmutation system but not necessarily for a system without transmutation, (2) reprocessing would be made even more expensive than normal recycling of uranium and plutonium because of the need to recover additional elements (e.g., neptunium, iodine, technetium), and (3) the need to operate a transmutation device with a lower breeding ratio than would be adopted for operation without transmutation. The most sensitive incremental cost factor at present is that of reprocessing spent LWR and other fuel to feed the S&T system.

Implications of Fuel Reprocessing Costs

The committee's review of cost data for reprocessing LWR spent fuel yielded an estimate of unit costs of $810 to $2,110/kg, depending on the type of financing and assuming construction in the United States (see Chapter 6). The Nuclear Energy Agency of the Organization for Economic Cooperation and Development (NEA/OECD) estimated that a reprocessing cost of $720/kg would result in a 14% increase in fuel-cycle cost of LWRS, as compared to a once-through fuel cycle, assuming government-type financing in Europe. The reprocessing cycle would add about 0.8 mill/kWh to the generating cost and about 1.5% in the total generating cost of wholesale electricity (i.e., busbar cost of electricity).

Studies of the actinide-burning fuel cycle supported by the U.S. Department of Energy have included cost estimates based on reprocessing facilities owned and financed by private capital. Consequently, the cost of reprocessing facilities, the fuel-cycle cost, and the cost of electricity would be greater than that estimated for government-owned facilities. To illustrate the importance of financing, we consider the effect on the NEA/OECD cost estimates for the LWR reprocessing fuel cycle resulting if reprocessing facilities were located in the United States and constructed and operated as a private venture with industrial financing. We adopt the estimated cost of $2,100/kg for a contemporary aqueous-technology reprocessing plant constructed in the United States (see Appendix J). The cost increment over the once-through fuel cycle would be about 4 mills/kWh, an increase of about 70% in the fuel-cycle cost and about 8% increase in the total generating cost of wholesale electricity.[5] At the present price of reprocessing services (about $1,000/kg—see Appendix J for details of a range of prices), the gross cost just for reprocessing 62,000 MgHM of LWR spent fuel would be $62 billion.[6] The total gross cost for recycle will be even greater when additional costs for fabricating recycle mixed-oxide (MOX) fuel are included.

It is predicted by some that the price of uranium will increase as resources are consumed and that by the second half of the next century the cost penalty for recycling may disappear. If this occurs, an economic benefit for recycling is expected to develop for breeder reactors. Even after the end of the next century, it would be expected that an economic benefit for a transmutation fuel cycle using an ALMR would develop.

The era of parity would be postponed if the future growth of nuclear power worldwide is less than expected, and it would arrive sooner if growth is more than expected. The deployment of an S&T system to improve waste disposal before an era of parity would need to be justified on the basis of benefits to nuclear waste disposal or to other parts of the nuclear fuel cycles.

There is also the possibility that the cost for reprocessing and transmutation could be used instead to improve waste forms for the once-through fuel cycle and the conservative design of the repository. This alternative has not been explored by this committee.

Pyroprocessing and Cost Perspective

The ALMR/IFR project estimates that it can achieve a twofold or more reduction in the cost of reprocessing spent LWR fuel using pyroprocessing technology compared to the use of aqueous separations technology. The committee believes that improvements in pyroprocessing may be possible but will have to be verified by research and development. Assuming the feasibility of pyroprocessing of spent LWR fuel, the design information for a commercial-scale reprocessing (IFR) facility needed to make cost estimates is not available.

Conclusion The excess cost for an S&T disposal system over once-through disposal for the 62,000 MgHM of LWR spent fuel is uncertain but is likely to be no less than $50 billion and easily could be over $100 billion if adopted in the United States. The additional cost of generating wholesale electricity could increase from 2 to 7% for a total increase of about $25 to over $80 billion.[7]

Findings Regarding Effects on Repository Capacity and Program

The Need to Dispose of Fission Products and Residual Wastes

All of the transmutation concepts require a geologic repository for at least some long-lived fission product radionuclides and for waste from some process losses and contaminated salvage materials.

Transmutation and Repository Capacity

Transmutation of the TRUs would reduce significantly the waste heat generation after a few hundred years in the

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[5] Fuel-cycle costs include the costs of ore, enrichment, fabrication, waste disposal and reprocessing if reprocessing occurs. It does not include the costs of the nuclear reactor and its operations.

[6] Should the actinide recovery lead to a simpler licensing process for the repository and the fuel reprocessing recover both the plutonium and uranium for possible electricity production, there would be a reduction in this cost. Also, if actinide recovery (and presumably elimination) proved to be necessary to allow the use of the Yucca Mountain repository, then the avoided costs of having to find another site would be large.

[7] The current charge of the repository program is approximately 1 mill/kWh or about $30 billion for the current fuel cycle.

repository, offering possibilities of increasing repository loading by a factor of four to five. But, transmutation is only one of several means (some of which do not involve S&T) of increasing the waste disposal capacity of a repository.

The repository loading can be increased without transmutation, such as by staggered loading, but the presence of actinides limits this increase to about 20%. In this method, alternate drifts and boreholes are filled over for a number of years; later, the remaining empty ones are filled (see Chapter 6). This approach is best suited for the period when strontium and cesium dominate the heating, i.e., over the first few centuries.

Some believe that a geologic repository sited at Yucca Mountain should be maintained hot in order to keep it dry. In this case, sustained high heat loading would be a benefit, and removal of the TRUs to reduce the heat loading would not be desirable.

In addition to the limit in repository size resulting from the technical issues, there is a statutory limit that could possibly be changed. Increase in repository capacity would increase the calculated long-term individual dose from that repository, although it would reduce the number of people that might receive doses from multiple repositories.

Conclusion Transmutation could increase the effective capacity of the first repository and thus delay the need for a second repository. In most cases, however, there are other ways to achieve the same end without using transmutation.

Findings Regarding Nontechnical Issues

Institutional and Public Policy Considerations

Embarking on any S&T system would pose difficult institutional and public policy problems. These include (1) the funding, operation, and overall management of a large and complex program of an extraordinarily long duration involving a linkage of commitments among many private and governmental organizations; and (2) the siting, licensing, and regulation of many major interdependent nuclear facilities. Public acceptance of such a system seems highly questionable under present conditions.

Legislative and Administrative Constraints

Creation of a S&T system would require major legislative and administrative steps to establish the policy basis for a national commitment to expenditures on the order of tens of billions of dollars needed to cover the net incremental cost of S&T until the increasing cost of uranium makes it economic. The operations would link numerous organizations over a period of many decades to centuries. Siting approval would require consent of federal, state, and local jurisdictions.

Financing

If private industry is to participate, plans, decisions, and expenditures to ensure the continuing integrity of the S&T systems have to be coordinated among and agreed upon by many private, federal, and, state organizations and agencies to ensure that such long-term commitments are prudent investments. In view of the history of nuclear power and commercial reprocessing in the United States, federal guarantees are likely to be necessary before private industry agrees to be involved with the construction and operation of the required S&T facilities.

Proliferation Issues

Widespread implementation of S&T systems could raise concerns on international proliferation risks. Since a number of the countries using nuclear power are committed to reprocessing, this may not be a decisive factor. It would, however, require the United States to change its policy against reprocessing.

Public Acceptance

If the current attitudes continue, siting a large number of nuclear facilities would likely raise considerable public opposition. There is no reliable basis for knowing whether an informed public would regard S&T as a significant and worthwhile improvement over the once-through LWR fuel cycle. The experience with nuclear programs of the past 20 years, however, strongly suggests that developing such public support will be difficult.

Conclusion The construction and operation of an S&T system would require, in addition to several new types of facilities, the resolution of major institutional, public policy, and public acceptance issues. For S&T to make a significant reduction in the amount and character of the radioactive waste slated for a geologic repository, a sustained, long-term national commitment would be necessary. The U.S. government would also have to accept the lead management and financial responsibility, with a cohesive national intent and commitment. The last two decades of U.S. government-led nuclear programs provide little confidence that such conditions can be established and maintained. Based on the experience of licensing recent power reactors, it is also highly questionable whether the degree of public support needed could be obtained now or in the near future.

Findings Regarding Research and Development

Scope for Research and Development

A research and development program in selected nuclear technologies, including pilot-scale demonstration of advanced separations, could serve a threefold purpose: (1) prepare the technical basis for alternative waste forms for the radionuclides in spent LWR fuel; (2) undergird more efficient future use of fissionable resources (e.g., using a breeder reactor), of which nuclear waste S&T could be an integral part; and (3) facilitate effective access to and influence on nuclear technology research and development programs being conducted in the rest of the world.

Conclusion A sustained but modest, carefully focused program of research and development over the next decade, emphasizing advanced separations technologies, could provide a sound basis for future decisions on nuclear waste S&T. Such a program does not now exist in the United States and needs to be developed. Following successful laboratory-scale development, engineering pilot-scale demonstration would be necessary.

Findings Regarding Defense Wastes

Of all the radioactive and mixed (hazardous and radioactive) wastes arising from the production of nuclear energy, weapons research, and production, the HLW stored in tanks is of the greatest importance.

The concentration of TRUs in most defense wastes is several orders of magnitude lower than in commercial HLW, and the total amount of TRUs are much smaller than in spent reactor fuel. For this reason it is not practical to transmute these wastes. However, since separations technology has an impact on the treatment and management of the defense wastes, the committee examined several treatment options in detail.

There are a number of ongoing programs for specific treatment of such wastes. The magnitude and diversity of the tank waste problems are greater at Hanford than at other DOE defense sites and are the cause of much local and national concern. For these reasons, the committee has treated Hanford as a case study of U.S. defense waste and has made specific conclusions and recommendations for the waste at Hanford.

Safety of Hanford Waste Storage Tanks

The safety of some of the Hanford tanks has been a major concern because they contain heterogeneous mixtures of gaseous hydrogen, organic compounds, ferrocyanides, sodium nitrate, and heat-generating radionuclides that are potentially reactive. The tanks that pose safety concerns have been identified. Characterization of their contents and mitigating measures are well under way.

Characterization of Hanford Tank Wastes

The present core sampling program for the heterogeneous contents of the Hanford tanks can be expedited but would give only unrepresentative analytical data for many tanks. Although appropriate sampling and analysis is necessary for safety concerns and some process planning, the data from production records for many tanks could be sufficient to start the removal and processing of their contents. Analysis of the resulting solutions and solids would yield representative data far more useful for further process development.

Retrieval, Separation and Treatment Potential for Hanford Tank Wastes

Leaving all the Hanford tank waste in place is not a viable option due to immediate and future safety problems and to public concerns. A promising conventional technique, caustic sludge washing, should remove more than 95% of the contents by dissolution and hydraulic disintegration of the solids. The solution removed would contain much of the ^{137}Cs and some of the ^{90}Sr, the removal of which would produce a waste that is approximately equivalent to the Nuclear Regulatory Commission Class C low-level waste category and contain 80-90% of the original tank solids after evaporation. The waste could then be stored. The remaining fraction of the retrieved waste, representing possibly 10% of the original waste volume, could be vitrified for geological disposal. Further separations, such as PUREX/TRUEX, could reduce the vitrified HLW to about 5% of the original volume. More extensive separations that would reduce the volume to 0.5% have been suggested. Such extensive advanced processing appears to be quite expensive and reliant on processes tested only on a laboratory scale. A research and development program tailored to the actual washed residues is under way to evaluate the technical and economic issues associated with the separation processes that could follow caustic sludge washing.

Conclusion Operations required to separate the Hanford tank wastes into a large-volume, low-activity portion destined for on-site disposal and a small-volume, high-activity portion destined for the repository are justified, primarily on economic grounds. At a minimum, justified operations include waste retrieval, sludge washing, and separation of key radionuclides from the large-volume portion. Further research and development on the processes and justification for additional separations involving the sludges is warranted.

For the 250,000 M^3 of Hanford tank waste, a relatively simple "sludge-wash" process should be implemented to reduce the volume of high-level waste to about 25,000 M^3. Research and development should continue on additional pretreatment (e.g., separations) options, and these should be implemented if they prove cost effective or are required to meet regulatory or institutional requirements.

The extensive further processing necessary to isolate the small amount (less than 1 Mg) of TRUs for transmutation would be unjustified.

PRINCIPAL RECOMMENDATIONS

The committee recommends the following:

- None of the S&T system concepts reviewed eliminates the need for a geologic repository. DOE should continue efforts to develop a geologic repository for spent LWR fuel.
- The current policy of using the once-through fuel cycle for commercial reactors, with disposal of the spent fuel as HLW, should be continued.
- Fuel retrievability should be extended to a reasonable time (on the order of 100 years) to avoid foreclosing alternative fuel strategies that may be in the national interest.
- Research and development should be conducted on selected topics to support the cost-effective future application of S&T of commercial spent fuel and separations for defense waste applications.

A sustained, but modest, and carefully focused program of research and development over the next decade could prepare the technical basis for advanced separation technology for the radionuclides in spent LWR fuel and for decisions on the possible applications of S&T as part of the more efficient future use of fissionable resources. The research and development effort should focus on the factors that strongly influence fuel-cycle economics, especially the costs of reprocessing spent LWR fuel, minimalization of long-lived radionuclides to secondary wastes in the reprocessing cycle, and on the need to minimize the possible increase in proliferation risks that could result from the commercial use of plutonium in recycle fuels.

Chapter One—

Introduction

The single most important challenge facing the nuclear field (commercial and defense) is what to do with the nuclear waste.

The Nuclear Waste Policy Act of 1982 commits the United States to geologic isolation as the best long-term solution to the final disposition of waste. Twelve years and billions of dollars later, there remain numerous institutional and technical questions concerning our ability to develop public confidence in full reliance on a geologic solution to the waste management problem. The result is the need to consider alternatives that either would stimulate progress towards a geologic solution in which this public has confidence, or to inspire other solutions that would reduce the dependency on any single approach to radioactive waste management. Concepts such as the separation and transmutation (S&T) of nuclear wastes that either eliminates or reduces their radioactive inventories are recognized alternatives. These alternatives were studied for many years prior to the Nuclear Waste Policy Act. With continuing difficulties, including the high costs, of demonstrating the long-term safety over thousands of years of geologic isolation, it is time to examine them with greater intensity. That is the purpose of this study. Many believe that nuclear energy is not an acceptable option until its waste products can be disposed of in a demonstrably acceptable manner.

Wastes are generated in the production of nuclear weapons and by the operation of the more than 100 nuclear power producing plants. The former are generally referred to as "defense wastes"; the latter as "light-water reactor (LWR) spent fuel." (Because there are potentially useful amounts of plutonium, uranium, and rare metals in LWR spent fuel, there is justification in considering LWR spent fuel as an energy and materials resource rather than waste.)

In the United States, most civilian nuclear power has been produced by the once-through LWR fuel cycle. The spent fuel is stored predominantly at the sites of the power reactors from which it has been discharged. This material cannot remain safely where it is in its present form for an indefinite period, but it might be stored at these locations long enough to reduce significantly the decay heat of the ^{137}Cs and ^{90}Sr radionuclides that have about 30-year half-lives. Defense wastes are located at Department of Energy (DOE) sites throughout the country. Many of the defense wastes are stored as liquids in underground tanks and so are not yet in a stable, well-contained form. This must be treated, or better controlled in place, to diminish their risk to the public.

A variety of approaches to achieving the goal of safe and ultimate disposal of nuclear waste are being considered. One of these involves "separations and transmutation" (S&T) to separate the hazardous long-lived radionclides from the wastes and transmute them either by fission or by neutron absorption to generate shorter-lived or stable isotopes. In general, it is considered that safe disposal for short times (times of the order of 1,000 years) will be much easier to achieve and to guarantee than long times (times of the order of 100,000 years or more).

COMMITTEE MISSION AND OPERATION

Committee Formation and Mandate

In 1991, the Secretary of Energy, Admiral James D. Watkins, Jr., requested the National Research Council to conduct a broad systems review of the application of separations technology and transmutation systems to radioactive waste disposal. To implement this request, in June 1991 the NRC formed a 19-member multidisciplinary committee on Separations Technology and Transmutation Systems (STATS) under the direction of the Board on Radioactive

Waste Management. The committee was assisted by a subcommittee on Separations, under the Board on Chemical Sciences and Technology, and a subcommittee on Transmutation, under the Energy Engineering Board. These subcommittees included 10 additional experts to support and report to the STATS committee. The expertise of the STATS committee and its subcommittees included chemical, nuclear, and fuel-cycle engineering, separation science, accelerator physics, radioactive and hazardous waste disposal technologies, exposure assessment, economics, public policy, regulatory policy and procedures, nuclear reactor safety, and plant operations. A third STATS subcommittee on Integration functioned until May 1992. Biographies of the committee members are given in Appendix M. The proposed statement of work of the STATS study sent to DOE in May 1991 is given in Appendix A.

The objective of the committee was to prepare a report covering its terms of reference as mutually agreed on between Admiral Watkins, Secretary of Energy, and Frank Press, Chairman, National Research Council, as elaborated in the Statement of Work given in Appendix A. In summary, STATS was requested to

- perform an independent review of both mature and developing separations technologies available in the United States and in other countries;
- evaluate the application of separations technology to DOE's radioactive defense waste management program;
- review the potential for application of separations and transmutation technologies to commercial spent nuclear fuel and potential impacts of successful waste separation and transmutation processes on the design and licensing of a deep geologic repository; this includes review and evaluation of potential life-cycle costs, benefits, ramifications, and program linkages between chemical separations and actinide or fission product transmutation in liquid metal reactors, accelerators, or other devices for the long-term management of high-level wastes; and
- recommend options that are economically and technically feasible and in compliance with all applicable regulations.

To initiate the study, an international workshop was held in January 1992 where the committee was briefed by a number of people representing a wide cross section of disciplines. To facilitate the work of the committee, selected members made site visits. Committee and subcommittee meetings were held as necessary (see Appendix B).

In May 1992, in response to a request by the Secretary of Energy, an interim report of the committee was submitted that summarized the committee's information at that time concerning actinide burning in advanced liquid metal reactors and the possible impacts on the repository program (see Appendix L).

Why Actinide Burning is Being Reconsidered

The resolution of the nuclear waste disposal problem is a crucial factor for the future of nuclear power in the United States. It is perceived to be one of the most important elements in establishing public confidence for continued or expanded nuclear electricity generation. Further, it plays a major role in the efforts to successfully manage and clean up the by-products from the nuclear weapons production complex. Consequently, any proposal for alternate approaches can easily capture the attention of numerous scientific and public constituencies. For both institutional and technical reasons, actinide burning is now being reconsidered.

Institutional Incentives

The apparent lack of progress on developing a nuclear waste repository has heightened public awareness and concern about achieving a solution to nuclear waste disposal. After two decades of federal programs and one decade after focused congressional action, an operating repository for civilian reactor wastes is still at least 15 years away. Cost estimates indicate that almost $10 billion will be expended before a decision to select a site and begin licensing actions will be made. Regulatory complexity and uncertainty is likewise affecting the progress toward and confidence about a final solution. Isolation standards are being reformulated and the associated regulatory structure is continuing to evolve. Finally, no significant progress has been made to establish broad-based political support for either a civilian waste repository or a monitored retrieval storage facility.

Public perception appears focused on the long-lived radioactive elements and those that do not occur naturally. For this reason, the actinides, especially plutonium, are of special concern. Consequently, any mechanism to reduce or eliminate these elements will draw attention as an alternative to the seemingly stalemated approach to nuclear waste disposal. Similarly, the complex regulatory requirements for disposal imply that any mechanism to simplify the licensing process or make the contents of a repository more benign would be attractive. Further, if the nuclear waste in a repository could be shown to pose a threat for a significantly reduced period of time, it is perceived by many that proof of ultimate safety would be easier to provide. Subsequent discussion in the report will indicate, however, that while actinides and their reduction may be a big factor in evaluating transmutation concepts, separations and transmutation of long-lived fission products may represent an equally significant concern.

Technical Incentives

Recent technical progress in several areas has been proposed as justification to reconsider transmutation. Repro cessing technology has advanced in many countries, offering the promise of highly efficient and selective separation operations. Likewise, the advance of robotics and remote handling gives support to cleaner, more efficient processing of radioactive materials. Advanced reactor designs, specifically liquid-metal reactors, which have the potential to consume actinides and incorporate integrated reprocessing facilities, are reaching a conceptual development and demonstration stage. Further, the possibility of neutron production, using high-energy accelerators, has been enhanced by recent advances in accelerator technology. Proponents argue that such neutron production can be used to drive subcritical reactor assemblies, providing high neutron fluxes capable of transmuting actinides and fission products while providing some safety advantages.

Scope of Study

The scope of the study was to prepare a reviewed report evaluating the relative effects, costs, and feasibility of employing separations and transmutation technologies in the Department of Energy's programs for managing (1) spent nuclear fuel from civilian power reactors, and (2) radioactive wastes from selected existing defense production reactor sites.

This final report of the STATS Committee is a peer-reviewed evaluation of the separations technology and transmutation systems application to radioactive waste management and disposal, with conclusions and recommendations.

The report covers in detail the current status and potential applications of separations and transmutation technologies and systems. These include critical reactors and subcritical assemblies driven by accelerators, in conjunction with aqueous and/or nonaqueous separation processes for separating actinides to be transmuted.

The report further examines whether practical implementation of feasible separations and transmutation (hereinafter referred to as S&T) concepts can contribute to a safer and more economical waste disposal method than what is now being considered. The report also assesses a time frame for such implementation.

Time-Frame

If there is a justification for the introduction of S&T, the time required for development and project implementation is important.

The development phase is envisaged by the various proponents of the S&T concepts to end by the first decade of the next century and the implementation phase to start thereafter. The report reviews whether these dates are realistic, taking into consideration the following important factors:

- achievable separation efficiencies and actinide reduction factors over time;
- consideration of whether the S&T systems are to be developed solely for waste management needs or also for power production;
- realistic assessment of the time required to develop the concepts to achieve practicality;
- health and safety issues;
- economics;
- policy and public acceptance considerations; and
- licensing and regulatory issues.

S&T Areas

Current and Future Waste Management Programs. Assessment of the current waste management programs, future needs, and associated issues can indicate whether the present fuel cycle (see Appendix C) needs any change. If changes are needed, it would be necessary to identify the reasons for and the timing of such changes. Answers to these questions lead the scope of this study to include: (1) separations-only and storage/disposal concepts; (2) integrated systems consisting of S&T; and (3) impacts, positive and negative, of S&T technologies on waste inventories, management and disposal over the entire fuel cycle.

Major considerations such as risk reductions, cost implications with cost-to-benefit analyses, and institutional feasibility and stability will be important for decision-making. Since most of the S&T proposals are based on concepts and paper studies, full details of the processes and systems are not available. The conclusions of the report are based on current national and, where relevant, international knowledge and status of S&T work.

Impact on National Energy Strategy. The National Energy Strategy (U.S. DOE, 1991/1992), projects that a substantial amount of new generating capacity—from 190 to more than 275 gigawatts—would be added through nuclear power between 1991 and 2010. It concludes that nuclear power is best utilized for "base-load" capacity. This strategy undoubtedly depends on the strategy's conservation actions.

A projected 195 to 290 gigawatts of nuclear capacity could be on line by the year 2030 if (1) favorable economics exist, (2) the public and the regulators deem the new plants to be safe, and (3) a solution to the disposal of nuclear waste can be found.

Reducing uncertainties, assuring safety, and promoting stability in the licensing process can lead to more predictable construction times and costs.

Implications of the nuclear energy component of the National Energy Strategy are important in determining S&T's role. In this report, two example cases, the declining power and the continuing constant power, are given to illustrate the effects of S & T.

Technologies

Separations technology employing PUREX has been used to obtain plutonium and uranium from production reactor fuel in the weapons program for many years from the beginning of the nuclear industry using the plutonium and uranium extraction (PUREX) aqueous processing of spent thermal reactor fuels. Although dozens of other promising separation methods have been studied in the laboratory and in pilot plants, including nonaqueous reprocessing, none has been developed to the same level for practical application as the PUREX process. The report reviews the existing PUREX process and other aqueous and nonaqueous processes under development, including studies on the chemistry of solvents.

The advanced liquid metal reactor (ALMR) program includes the integral fast reactor (IFR) under development by Argonne National Laboratory (ANL) and the Power Reactor, Innovative, Small Module (PRISM) modular reactor concept under development by General Electric (GE). The primary aim of the ALMR is power production as a breeder. However, the ALMR can be modified for transmutation of transuranic wastes (TRU). The LWRs could be used as plutonium burners, as transmuters of actinides, and transmutation of fission products ^{99}TC and ^{129}I. Existing LWRs are in an advanced stage of development, the proposed next generation being advanced light-water reactors (ALWRs). The Clean Use of Reactor Energy (CURE) concept proposed by Westinghouse Hanford Co. envisions the use of both ALMRs and LWRs in a system transmuting actinides as well as fission products. Finally, Brookhaven National Laboratory (BNL) proposes the modification of their conceptual space particle bed reactor (PBR) to recycle uranium as well as unburned plutonium. The information on these latter two concepts is so meager that no analysis of their performance has been possible.

High neutron fluxes can be attained by the use of accelerators for fast burn-up of the actinides and certain fission products. Los Alamos National Laboratory (LANL) and BNL have undertaken several conceptual studies for achieving accelerator-based transmutation. The accelerator scheme envisions the production of electricity—part of it used to power the accelerator and the rest for sale—an important element in paying at least part of the cost of transmutation.

Development of the concepts described above to achieve practical S&T realities—economic and technical factors taken into account—will have to overcome many problems. The report highlights and analyzes these problem areas and assesses the options that have potential for further development.

BACKGROUND ON SPENT REACTOR FUEL AND HIGH-LEVEL WASTE DISPOSAL

History

The final disposal of spent reactor fuel and high-level radioactive waste (HLW) from nuclear reactors in the United States was initially addressed early in the development of nuclear power. It was recognized that the waste from civilian power reactors and atomic energy defense activities was sufficiently hazardous as to require long-term isolation from human populations. In a 1957 study for the U.S. Atomic Energy Commission (AEC), the National Academy of Sciences recommended that high-level waste be buried in deep underground repositories and suggested bedded salt deposits as a likely host rock. Generic engineering tests were performed in the mid-1960s to see the effects of emplacement of spent fuel on salt and vice versa in a salt mine near Lyons, Kansas. Shortly thereafter, conceptual designs were developed for a repository at that location. The high-level waste program at that time focused on technical issues, principally the interactions between waste forms and the host rock and the geologic and hydrologic integrity of the site. However, it soon became clear that public and institutional involvement would play an important role in the progress of the high-level waste program. In 1972, after encountering some technical difficulties and institutional issues, the AEC abandoned the Lyons' project and sought to identify another site in salt deposits. In addition, the AEC announced a program to employ retrievable surface storage as an interim measure until a repository could be developed.

Site investigations began in southeastern New Mexico in the early 1970s to identify potential repository locations in the western part of the Permian basin. In 1975, the Energy Research and Development Administration (ERDA) identified a location in salt approximately 30 miles east of Carlsbad, New Mexico, that ultimately became the site for the Waste Isolation Pilot Plant (WIPP). At the same time, ERDA gave up plans for early construction of a retrievable surface storage facility. Evaluation of other locations around the United States for a nuclear waste repository began in 1976. In 1980, ERDA's successor agency, the Department of Energy, issued a generic environmental impact statement that examined numerous strategies, including transmutation of radionuclides to a more benign form, and selected mined geologic disposal as the approach for waste isolation in the United States. This action and a concurrent government policy also confirmed that spent fuel from civilian nuclear power reactors would not be reprocessed but disposed of directly in a repository (The Nuclear Waste Policy Act was silent on the question of spent-fuel reprocessed waste disposal). The following year the U.S. Con gress established the WIPP project in New Mexico as a demonstration project for disposal of defense transuranic waste. Congress precluded licensing of WIPP by the Nuclear Regulatory Commission (NRC) and did not require WIPP to demonstrate compliance with Environmental Protection Agency (EPA) standards prior to emplacement of waste.

Faced with significant controversy and regional concern, in 1982 Congress enacted PL 97-425, the Nuclear Waste Policy Act (NWPA). The act provided the basis to select and develop sites for two nuclear waste repositories sufficient to accommodate the high-level radioactive waste produced by both the civilian and defense sectors. The NWPA also provided for the establishment by the EPA of standards for protection of the general environment from off-site releases of radioactive material in repositories. In the case of civilian HLW and spent fuel, the NRC's role is as the repository licensing authority in compliance with these standards. The NWPA also mandated that the fuel costs of the HLW management program be paid through a nuclear waste fee. A fee of one mil ($0.001) per kilowatt hour of nuclear-generated electricity is paid into the Nuclear Waste Fund and currently provides about $800 million a year (One mil per kilowatt is the initial value of the fee that is required to be charged as necessary to ensure full cost recovery). Total income and earnings to the Fund since its inception in 1983 is approximately $6 billion through 1992 and $8 billion through 1994. While this fund is generated entirely by ratepayers using nuclear electricity, and may only be used to support storage and disposal activities specified in the NWPA, it is still subject to congressional appropriations and deficit controls to establish annual allowable expenditures for the program. Through 1992, approximately $3.5 billion has been spent from the Nuclear Waste Fund. For the disposal of defense HLW, the NWPA specifies that fees be paid by the federal government via congressional appropriation.

In 1983, the National Academy of Sciences completed a system study on the geologic disposal of HLW that reviewed alternative technologies available for the isolation of radioactive waste in mined geologic repositories, evaluated the need for and possible performance benefits from those technologies, and identified appropriate technical criteria for selecting the appropriate technology.

In 1983, the NRC issued regulations (10CFR60) to guide its licensing process, and in 1985 the EPA promulgated the environmental standards (40CFR191). In 1987, the EPA standard was remanded by a U.S. federal court, and the EPA was required to evaluate and reissue a revised standard addressing the concerns of the court on the containment and isolation provisions in the EPA standard. In 1992 Congress required the EPA to develop a new standard for Yucca Mountain based on recommendations from a study by the National Academy of Sciences. In addition, the WIPP Land Withdrawal Act directed EPA to promulgate a standard specifically for WIPP and gave EPA authority for its implementation.

In 1985, President Reagan concluded that defense HLW would be disposed of in the repository being developed for civilian waste. During this same time frame, and consistent with the schedule laid out in the NWPA, DOE concluded that a monitored retrievable storage (MRS) facility was required for spent fuel from commercial nuclear power reactors. This facility would provide temporary storage and subsequent shipment to a repository.

The following year, three candidate sites were recommended to be fully characterized for potential use as the nation's first HLW repository. These sites were located in Hanford, Washington; Yucca Mountain, Nevada; and Deaf Smith County, Texas. All three sites were to be characterized in parallel and a preferred site identified for repository development.

In 1987, after significant public and institutional controversy, and faced with significant costs for characterization, Congress amended the NWPA. The Nuclear Waste Policy Amendments Act (NWPAA) eliminated the program to identify a site for a second repository, pending reconsideration in 2007, and provided for the characterization for only one site, Yucca Mountain, Nevada, for the first repository. The NWPAA also allowed for an MRS facility but tied its development and construction to that of the first repository.

Current Waste Disposal Situation

Spent Reactor Fuel

After significant institutional and political controversy, the Yucca Mountain Project is proceeding with the characterization of the volcanic tuff for development as a repository. The project schedule has a goal of submitting a license application to the NRC in 2001. Allowing 3 years for licensing and 6 years for construction, operation of the repository is currently scheduled to begin in 2010. The repository at Yucca Mountain would contain a total of 70,000 metric tons uranium (MTU) equivalent of nuclear waste, of which about 62,000 MTU would be spent fuel from civilian power reactors. This capacity is the statutory limit imposed by the NWPA and does not necessarily represent physical limits at the Yucca Mountain site. This spent-fuel content for Yucca Mountain is approximately equal to what will be discharged by U.S. reactors through the year 2010. The projected discharge of spent fuel from current reactors in the U.S. during their lifetime (by the year 2040) will be about 90,000 MTU, necessitating a second repository or congressional action to increase the limit on capacity in the first repository. Since the TRU content of spent fuel is approximately 1% by mass, 900 MTU of TRU will be generated by currently operating reactors, with approximately 600 MTU produced by the year 2010.

The repository program is encountering considerable controversy. The characterization of the Yucca Mountain site does not enjoy political support in the state of Nevada. Program costs have risen significantly, and the program has been delayed more than a decade in light of the apparent complexity of characterizing a repository consistent with NRC licensing requirements. No firm estimates are available of the cost of characterization, which is expected to exceed $5 billion before a license application can be submitted. For its part, the MRS program will also require securing a site in the face of probable political opposition. Further, the development of the MRS is by statute linked directly to progress on the repository; it is therefore likely that an MRS cannot be brought into operation on a schedule to support DOE's commitment to begin accepting spent fuel in 1998.

These factors have seriously eroded public and institutional confidence in the management and potential success of DOE's civilian radioactive waste management program. Concern has been expressed repeatedly with regard to an overly complex management system that places too little emphasis on the scientific and engineering evaluations to effectively determine the potential of the Yucca Mountain site on a timely basis. This apparent lack of progress and significant cost give rise to efforts to identify alternatives or simplifications to the development of a nuclear waste repository.

Lack of progress in the development of nuclear waste repositories has also been attributed, in part, to the complexity of the environmental standards and the difficulty in demonstrating compliance with these standards. Concerns over the ability to comply with the standards and the apparently overstructured, inflexible programs being implemented by DOE resulted in strong recommendations by Congress that the standards, approach, and program be reevaluated. These concerns were legislatively addressed in 1992 by congressional requirements (PL 102-486) that public health and safety standards be promulgated by the Administrator of the Environmental Protection Agency for a repository at the Yucca Mountain site. These standards must be based on findings and recommendations of the National Academy of Sciences. The standards were required to address the maximum annual effective dose equivalent to individual members of the public. The National Academy of Sciences was asked to provide findings and recommendations that addressed the appropriateness of a standard based on dose to an individual, impact of postclosure institutional control on repository risk, and the validity of scientific predictions regarding the probability of human intrusion over a 10,000-year period. Requirements were also place on the U.S. NRC to modify its technical requirements and criteria to be consistent with the new standards of the EPA. The legislation provided that the National Academy of Sciences committee issue a report. The National Academy of Sciences report entitled *Technical Bases for Yucca Mountain Standards* was issued in 1995.

Defense High-Level Waste

The production of nuclear weapons and naval propulsion systems has resulted in an accumulation of radioactive wastes at several locations around the United States. These wastes are typically classified as high-level radioactive waste (HLW), transuranic wastes (TRU), low-level radioactive wastes (LLW), and, more recently, mixed wastes. Liquid HLWs are the direct products resulting from the reprocessing of spent reactor fuel and irradiated targets. As generated, the wastes are highly acidic, but almost all of them have been neutralized with sodium hydroxide to prevent unacceptable corrosion of mild steel tanks. The wastes are stored primarily in large metal tanks containing various mixtures of liquids, sludges, salt cake, and solids (calcines). The locations and approximate amounts of HLWs in storage (U.S. DOE, 1991) are as follows:

Savannah River Site	132,000 m^3
Hanford Site	254,000 m^3
Idaho Chemical Processing Site	12,000 m^3
West Valley Demonstration Project	1,230 m^3

The composition of wastes at each site varies as a result of the specific processing and storage techniques used. The Idaho site, for example, has acidic liquids stored in tanks and a large volume of waste converted into a solid (calcine). The West Valley project has relatively smaller volumes of alkaline and acidic wastes that are intended for subsequent processing into a glass waste form for disposal. At Savannah River, significant quantities of wastes have been generated from the application of the PUREX process to fuel and targets from production reactors. A vitrification facility to convert these HLWs into a stable glass form is in final stages of completion.

The largest quantity of HLW is stored in 149 single shell tanks and 28 double-shell tanks at the Hanford site. These wastes, while larger in volume than those at Savannah River, typically contain less radioactivity. This is largely due to the removal of strontium and cesium into capsules that contain almost the same amount of radioactivity (170×10^6 Ci[1]) as the entire contents of the tanks. The chemical separation processes at Hanford have resulted in approxi

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[1] Throughout this report, the term Curie (Ci) is used as the unit of radioactivity. The SI unit of radioactivity is the becquerel (Bq). 1 Ci = 3.7×10^{10} Bq.

mately 1 Mg of TRU elements in the HLWs and approximately 2,000 Mg of uranium. The wastes at the Hanford site have been chosen as the reference case for application of transmutation and advanced separations to defense HLWs in this study.

TRU wastes, primarily solid, are the other significant quantity of defense wastes requiring geologic disposal. These wastes contain TRU elements in quantities greater than 100 nCi/g of total waste material. Prior to 1970 when TRU wastes were defined as those containing >10 nCi/g of TRU, these wastes were buried in surface landfills at DOE sites. Much of these buried wastes may actually be below the 100 nCi/g level for TRU waste. Since 1970, the TRU wastes were stored retrievably to be subsequently disposed of in the WIPP facility. In 1990, approximately 191,000 m^3 of buried TRU containing approximately 800 kg of transuranic elements was located at Hanford, Idaho National Engineering Laboratory, Los Alamos National Laboratory, Oak Ridge National Laboratory and Savannah River Site.[2] At that time, 61,000 m^3 containing approximately 2,200 kg of TRU elements was retrievably stored at these sites with over one-half of the total at Idaho National Engineering Laboratory. In addition to buried and stored TRU waste, it is estimated that as much as 1,800,000 m^3 of potentially contaminated soil is present at these sites. In addition, residues from Rocky Flats and remotely handled TRU waste, while smaller in volume, contain approximately 3,000 kg of TRU elements. Consequently, the transuranic element content of all categories of TRU wastes far exceeds that of the tanks at Hanford.

TRU wastes are extremely varied in composition, typically consisting of drums and boxes containing organic materials (papers, clothing, plastics), sludges, and contaminated laboratory or production equipment from nuclear materials processing. This nonhomogeneity and diversity in composition make TRU wastes unlikely candidates for advanced separations or transmutations. In fact, many of the concerns about safe disposal of TRU wastes are due to its chemical form and not its content of radioactivity; consequently, processing to modify the waste form is much more likely to result in effective containment in a repository than S&T of its relatively small content of TRU elements. Consequently, this study does not attempt to evaluate the application of S&T to defense TRU wastes.

Today, the proposed repository for defense TRU wastes, WIPP, has been completely constructed and awaits the results of a program to evaluate compliance with the EPA standard and a test program to obtain additional information on waste interactions. The program is expected to last several years. If the repository is found to comply with isolation standards of the EPA, disposal operations could begin in the second half of this decade. Agreement on the process to demonstrate compliance and the conduct of experiments using radioactive waters will need to be established. It is likely that continued interaction between DOE, federal and state oversight agencies, and public interest groups could mean additional delays for the program and ultimate operations.

A major step for WIPP was completed in 1992 when land withdrawal legislation was passed by Congress (PL 102-579), allowing DOE, after completing several prerequisite actions with the EPA, the National Academy of Sciences, and the state of New Mexico, to conduct the final stage of demonstrating compliance for the WIPP. (DOE subsequently decided to conduct the necessary tests above ground.) This legislation also requires the EPA to develop a disposal standard for WIPP, independent of the standard to be developed for Yucca Mountain.

Current Policies Governing High-Level Waste Management

The committee has reviewed two broad topics for their implications for waste management: (1) the S&T of certain specific radionuclides from spent fuel; and (2) the separation of HLW produced in weapons research, development, and production. Although not explicitly prohibited by law or regulation, the development and use of a system for transmutation would represent a radical change in current public policy and fuel-management plans. However, current policy does call for the separation of defense HLW before processing it for disposal. Changes in how and to what extent separation is used will require only a modification of current policy and plans.

Commercial Spent Fuel

Policy. The United States has a set of formal policies concerning the management and disposal of commercial spent fuel embodied in legislation, executive orders, and regulation. In particular, two major laws govern commercial spent-fuel management: the Nuclear Waste Policy Act of 1982 (PL 97-425) and the Nuclear Waste Policy Amendments Act of 1987 (PL 100-203). The 1982 Act was based on a broad consensus about some issues and on compromises concerning others. The 1987 amendments significantly revised the 1982 Act. Passage of each act required considerable compromise and expenditure of political effort. Congress will not lightly alter the basic agreements that were reached through the difficult negotiation required to balance competing interests.

Reprocessing. The first U.S. nuclear power plants were designed and built under the assumption that spent fuel

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[2] 800 kg of TRU contains predominantly ^{239}Pu with a specific radioactivity of 61 curies/kgm.

would be reprocessed, that the recovered plutonium and some other actinides would be used in fuel for light-water reactors, and that the resulting HLW would be solidified and then disposed. President Carter, however, delayed reprocessing because of concern about nuclear weapons proliferation. President Reagan removed the prohibition, declaring that reprocessing was the responsibility of private industry to develop without government assistance. Utilities now store spent fuel at power plants and expect to dispose of its as unreprocessed waste.

Interim storage. The NWPAA authorized the siting, construction, and operation of a monitored retrievable storage facility (MRS) for the interim storage of commercial spent fuel, subject to certain conditions linking MRS development to repository development: DOE cannot select a site for an MRS before the Secretary of Energy has recommended approval of a site for the development of a repository; the license for MRS must contain conditions that allow construction and operation of the MRS to continue only when repository construction and operation is proceeding. Total capacity of the MRS is limited to 15,000 tons.

The NWPAA also established the Office of the Nuclear Waste Negotiator to negotiate agreements with states or Indian tribes willing to host a repository or an MRS. Such an agreement could contain different conditions than those imposed on a DOE-sited facility. The term of the Office of the Nuclear Waste Negotiator has now expired, and the Office was not reauthorized by Congress.

Ownership of spent fuel. The federal government will eventually own and be responsible for the disposal of commercial spent fuel. The NWPA directed utilities to levy fees on electricity generated by nuclear power and to pay those fees into the federal Nuclear Waste Fund to be used to develop and operate a repository. In return, the NWPA directed the federal government to accept ownership of spent fuel "beginning not later than January 31, 1998" and to start accepting the waste as a repository is available.

Repository. Both acts establish building a repository for the disposal of HLW as a top government priority for HLW management. Some people support this policy because they believe that those who benefitted from the processes that generated the waste (that is, those who used the electricity) must dispose of the resulting waste. Others support the policy because they believe that the survival of nuclear power depends on demonstrating the ability to dispose of spent fuel.

The NWPA of 1982 limits the amount of waste placed initially in a first repository. This limitation was imposed as a matter of policy, rather than on the basis of technical considerations. Only the equivalent of 70,000 metric tons of heavy metal in spent fuel and/or in solidified HLW can be emplaced in a first repository before a second repository is operating. Under current plans, 7,000 metric tons of heavy metal equivalent of solidified HLW from the weapons program is to be disposed of in the civilian repository, leaving the balance for commercial spent fuel. The NWPA delays congressional consideration of the need for a second repository until after the year 2007.

According to Section 122 of the NWPAA, spent fuel placed in a repository must be retrievable during an appropriate period of operation of the facility for "any reason pertaining to the public health and safety, or the environment, or for the purpose of permitting the recovery of the economically valuable contents" of the spent fuel. NRC regulations require that provisions must be made to enable the spent fuel to be retrieved on a reasonable schedule, starting any time up to 50 years after waste emplacement begins.

Transmutation proposals. All proposals for transmuting commercial spent fuel require reprocessing as a first step. Some assume that reprocessing would occur initially to recover radionuclides for producing electricity and that additional transmutation for waste management and disposal purposes would occur as an auxiliary process. These proposals require that elaborate new reactor or accelerator systems be developed and operated, most for many decades. Such operations would result in new and varied waste streams. As discussed in Chapter 5, these proposals have far-reaching implications for current policies about the ownership of spent fuel and the fuel resources in it; responsibility and time duration for spent-fuel storage; the forms of the waste to be eventually disposed of and the implications for repository capacity and design; and the development, regulation, and financing of new technology for transmutation systems and their operation over decades. At present, there are no affirmative policies that support the extensive research, development, and capital investment required to develop a transmutation system.

Defense High-Level Waste

For many years the country's nuclear weapons facilities were largely closed to public review and outside regulation. However, increased public concern about environmental issues, passage of state and federal laws extending environmental regulation and oversight to government activities, and court rulings eventually led the federal government to acknowledge significant legal obligations to comply with environmental regulations and standards.

The legal basis for policies for managing and disposing of HLW in tanks at Hanford is contained in the Hanford Federal Facility Agreement and several current and planned environmental impact statements. The Washington State

Department of Ecology, the U. S. Environmental Protection Agency, and the U.S. Department of Energy signed the Hanford Federal Facility Agreement and Consent Order in 1989 (referred to as the "Tri-Party Agreement"). The agreement is based on provisions of Executive Order 12580, Washington State Hazardous Waste Management Act, and several federal laws: the Comprehensive Environmental Response, Compensation, and Liability Act, as amended by the Superfund Amendments and Reauthorization Act of 1986; the Resource Conservation and Recovery Act, as amended by the Hazardous and Solid Waste Amendments of 1984; and the Atomic Energy Act of 1954, as amended. The agreement may be amended by unanimous agreement of the signatories. An action plan delineates actions to be taken, provides a schedule, and specifies procedures for modifying or amending the action plan.

Under the 1989 Tri-Party Agreement, as amended until January 1994, tank waste is to be separated. The high activity fractions are to be solidified as glass and stored until disposal in a repository. The low activity fraction was originally destined to be immobilized in grout and stored on site; in 1993, the parties agreed to change the waste form to glass. In 1992, preliminary site preparation began for a waste vitrification plant at Hanford, but work was halted under an agreement to consider a revised technical strategy for Hanford. In 1993, the three parties agreed to suspend, until the year 2002, the start of construction of the high activity fraction vitrification plant, and agreed to begin construction of the low activity fraction vitrification plant in 1997.

ORGANIZATION OF THE REPORT

The report is organized in three parts: an Executive Summary, a main report of six chapters, and 16 appendices. The Executive Summary is a stand-alone document that gives the findings, the recommendations, and the conclusions of the STATS Committee. The Executive Summary also, in most cases, explains the basis on which the conclusions, recommendations, and findings were reached. The complete technical justification is not given in the Executive Summary. The detailed technical analysis appears either in the main report or in one of the 16 appendices.

The first chapter of the report gives the scope and objectives of the study and a brief history of U.S. efforts in the area of high level radioactive wastes from the late 1950s to the present. It also describes the organization of the report. Chapter 2 discusses the nature of the radioactivity in spent power reactor fuel. It reviews the radionuclides of most concern and looks at the potential public exposure from current and some proposed fuel cycles. This chapter also gives a description of the three Separations and Transmutation (S&T) scenarios.

Chapter 3 looks in detail at the separations technology that is available and could be potentially available for the separations required in the S&T approach. Chapter 4 looks in detail at the three candidates' concepts for carrying out the transmutation step in this approach. In particular, calculations are presented that show how long it requires to accomplish a specified level of burnup of the radionuclides of concern.

Chapter 5 looks at the problems of dealing with defense high level waste, which are usually liquids containing fission products with relatively little heavy metal. The particular case of the waste stored in tanks at the Hanford reservation are reviewed for applications of S&T to that particular problem.

Finally, Chapter 6 looks at a number of technical and societal issues that are involved in the adoption of any of the S&T systems proposed.

The appendices give more detailed technical information on the subjects identified in their titles. Because acronyms, abbreviations, and technical jargon are widely used in this technology, the reader may find appendices N and O useful.

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Chapter Two—

Summaries of S&T Proposals and Radionuclides Release Scenarios in Repository

This chapter gives a brief overview of the source and characteristics of radionuclides in spent fuel, summaries of Separation and Transmutation (S&T) proposals being considered for commercial spent-fuel reprocessing and defense high-level waste treatment, and details of the performance of the radionuclides involved in a repository.

SOURCE AND CHARACTERISTICS OF RADIONUCLIDES IN SPENT FUEL

Radioactivity Produced by Reactors

After a nuclear reactor has been run at full power for a few months a large number of new radioactive species are produced. One of the nuclear reactions responsible for this radioactivity production is the neutron, gamma reaction often written (n, γ). In this reaction the target nucleus captures a neutron and emits one or more γ-rays. The target nucleus increases its mass number by one but does not change its atomic number. An example of such a reaction is the capture of a neutron by ^{238}U to produce ^{239}U plus some γ-rays. The product of the (n, γ) may be stable or radioactive. The transuranic isotopes shown in Figure 2-1 exhibit three forms of instability: α-decay, β-decay, and spontaneous fission (SF). None of the nuclides of atomic number greater than 92 are stable. It is also interesting to notice that the (n, γ) reaction can repeat itself and form a series of (n, γ) steps such as exhibited by Pu going from ^{238}Pu to ^{246}Pu. Decay by the emission of an ordinary negative electron called β-decay results in an increase of one in the atomic number. Note that the light-water reactor (LWR) starts with only ^{235}U and ^{238}U in the chemical compound uranium dioxide as fuel, and after a substantial run at power will have produced all 51 of the nuclear species shown in Figure 2-1, although those heavier than ^{244}Cm are present in amounts so small as to be negligible. These 51 species are called the actinides and are of particular interest to the STATS study. The actinides form one important component of the spent fuel produced in LWRs. Since the actinides other than uranium are all produced by nuclear reactions in the fuel, all of them are born inside the uranium dioxide fuel. A second type of reaction that can occur is neutron capture that leads to the fissioning of the target nuclide. In the fission process the capture of a neutron imparts enough energy to the struck nucleus to cause it to break apart into pieces called fission products. During the process, γ-rays and energetic neutrons are given off. Some of the actinides will fission upon capture of a low-energy neutron, e.g., ^{239}Pu; almost all will fission if the incident neutron has an energy above 1 MeV. The products of fission are neutron rich and almost all are unstable against β-decay. These fissions will almost all take place inside the uranium dioxide fuel. There are several hundred different fission products produced by these reactions which cause the spent reactor fuel to be highly radioactive and generate substantial amounts of decay heat. Just after shut down the decay heat from nuclear fuel in the reaction will still produce heat at about 7% of the full power output.

Another source of spent fuel radioactivity will be from a few actinides that decay by spontaneous fission, that is, they undergo fission without a neutron capture or any other source of energy being required. They differ from neutron-induced fission in that neutron-induced fission stops when the chain reaction stops but spontaneous fission continues to decay away at its characteristic half-life. Spontaneous fission neutrons can present shielding problems during reprocessing and fuel fabrication.

Neutron activation products are also generated when nuclear fuel is used to produce power. These are radioactive species formed by neutron bombardment of non-fuel materials such as fuel cladding and other structures inside the reactor. An example of such a case is that trace amounts of nitrogen in the fuel matrix produce ^{14}C, a long-lived activation product of concern in some waste disposal circumstances.

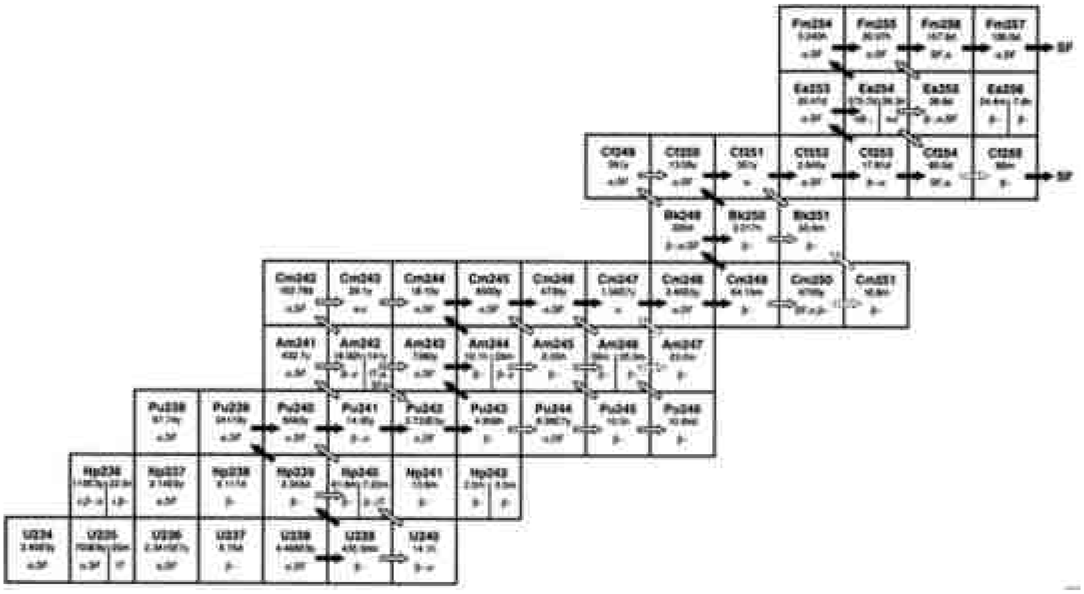

FIGURE 2-1 Neutron capture paths of importance for actinides.

The actinides, fission products, and activation products taken together are the radioactive waste that must be sequestered in some way so that they do not enter the biosphere in unacceptable concentrations. Today, these three radionuclide groups are contained in the uranium dioxide fuel matrix or structural materials of spent fuel assemblies. Almost all of the spent fuel is stored in pools at the reactor sites. Uranium dioxide is a ceramic material that effectively contains radioactive elements under most circumstances. However, storage of spent fuel at the reactor in cooling pools is considered only a temporary solution (less than a century). It is possible and legal to store the spent fuel out of pool up to 60-70 years. After that, a permanent solution will be required. The current government plan is to place the spent fuel, following encapsulation, deep underground in a stable geological formation.

All nuclides have some propensity for absorbing neutrons that changes them into a different species with different nuclear properties. This process is called transmutation. The goal of transmutation for waste management purposes is to convert a long-lived radionuclide that is potentially troublesome at a waste disposal site to a shorter-lived or stable nuclide by exposing the troublesome nuclide to a high flux of neutrons for a sustained time. This has the effect of reducing the long-term toxicity of the waste because most of the waste constituents would then decay to a non-radioactive nuclide in a short time. Since some of the nuclides in the spent fuel will produce unwanted nuclides if bombarded with neutrons, it becomes essential to separate carefully the nuclides before the neutron bombardment so that only those that produce the desired outcome will be included among those exposed to the neutron flux. This is the basic premise of the S&T method of waste disposal. There are several problems that make this more difficult than it might seem. First, in many cases a fraction of the neutron captures produce another long-lived radionuclide, which does little to achieve the transmutation goal stated above. A second problem is that the source of the neutrons is a nuclear reactor of some kind. Thus, as that reactor operates it will produce more of the same nuclides that are being destroyed, so the observed reduction will be the net of destruction minus production.

Toxicity Considerations and Isotopes of Concern

Toxicity Index

There are 200 to 300 different radioactive species generated by the operation of a nuclear reactor, primarily as a result of neutron capture and neutron-induced fission. The adverse impact of the various radionuclides varies because of the differences in the chemical behavior in the body of, and the radiations emitted by, the radionuclides. To account for differences in the extent to which humans are hypothesized to be damaged by exposure to radionuclides it is common to introduce the concepts of ingestion and inhalation toxicity indices. The former measures the toxicity of exposure if one ingests the waste and the latter if one inhales the waste. The toxicity index is expressed as:

$$TI_k = \frac{\sum_i \lambda_i N_i}{C_{ik}}$$

where:

TI_k = The volume of water (or air) required to dilute a radionuclide to the maximum permissible concentration for pathway *k*, m^3 of water (or air)/mass or volume of radioactive material.

$\lambda_i N_i$ = the product of the decay constant and number of atoms of radionuclide *i* which yields the activity (radioactive decay rate), curies (becquerels).

C_{ik} = Maximum permissible concentration of radionuclide *i* for pathway *k*, curies (becquerels)/m^3 of water (or air).

The toxicity index is a measure of the potential for adverse radiological impacts to biological organisms associated with exposure to a defined mixture of radionuclides. The toxicity index is *not* a measure of risk. The risk from a mixture of radionuclides is a function of the potential impacts from exposure as well as the likelihood that exposure will take place. For example, ingestion or inhalation of the contents of a HLW container might produce grave results so the material is considered a highly toxic substance. However, if it is in a well-selected and well-designed repository so that the extent of human exposure is very small it represents a very low risk.

The primary focus of the toxicity of radionuclides in this report is related to waste disposal in a geologic repository. The most common release and exposure mechanisms from a repository involve ground water contacting the waste form followed by slow dissolution, transport of radionuclides to the accessible environment, distribution in the biosphere, and eventual uptake from food and water. As a result, the ingestion toxicity index is of particular importance. In Figure 2-2 the toxicity from ingestion as a function of decay time is shown for a number of radionuclides contained in spent PWR fuel. The activities of the radionuclides in the spent fuel were taken from (Croff, 1980a) and the maximum permissible concentrations were taken from Appendix B, Table 2, Column 2 of 10CFR20 [10CFR20 1991] which are calculated to produce an effective dose equivalent of 50 millirem/yr[1] .

Isotopes of Principal Concern

Although hundreds of isotopes are present in spent fuel or wastes derived from them only a few of them are important in HLW and spent fuel disposal. A key reason for this is that the NRC regulations in 10CFR60 require that the containment of the waste constituents of a HLW package be substantially complete for a period in the 300-to 1,000-year range after permanent closure of the geologic repository. After 20 half-lives the radioactivity will be reduced by a factor of 2^{20} ` 10^6. We assume this requirement is implemented with a high degree of confidence and materials with half-lives less than 50 years (1,000 yr/20) will have decayed to negligible levels before a significant number of packages failed. Thus, for the purposes of ascertaining the health risk posed by the repository, radionuclides having half-lives less than 50 years are ignored.

By eliminating radionuclides having half-lives of less than 50 years as contributors to the repository health risk, there are two classes of radionuclides that are of concern: medium-lived fission products that substantially affect repository performance and long-lived radionuclides that are produced in significant amounts.

Fission Products. There are two fission products that are a principal concern because of their substantial thermal impact on the repository as opposed to posing a potential health risk: ^{90}Sr and ^{137}Cs. These two radionuclides, which could pose a health risk in an early intrusion scenario, are dominant contributors to the heat released by spent fuel or HLW during at least the first several decades (see Appendix Figure G-5). This heat is a major consideration in repository design and a significant issue in determining the performance of the repository. Cesium-137 is also a major source of penetrating radiation emitted by spent fuel and HLW during the first several decades which is a major consideration in handling these wastes. Some of the properties of these two radionuclides are summarized in Table 2-1. The combination of being medium-lived and having relatively small cross sections makes transmutation impractical for these radionuclides. A number of other methods have been proposed to eliminate this problem and are explained above in the section *Impact of S&T on Waste Repository* .

Two other fission products are of principal concern because of their potential contribution to health risk: ^{99}Tc and ^{129}I. They are of principal concern because they are long-lived, produced in significant amounts in the fission process (see Table 2-1), generally soluble under geologic conditions, and migrate relatively quickly under common ground water conditions.

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[1] Throughout this report, the term Rem (R) is used as the unit of dose equivalent. The SI unit of dose equivalent is the Sievert (Sv). 1 Sv - 100 R.

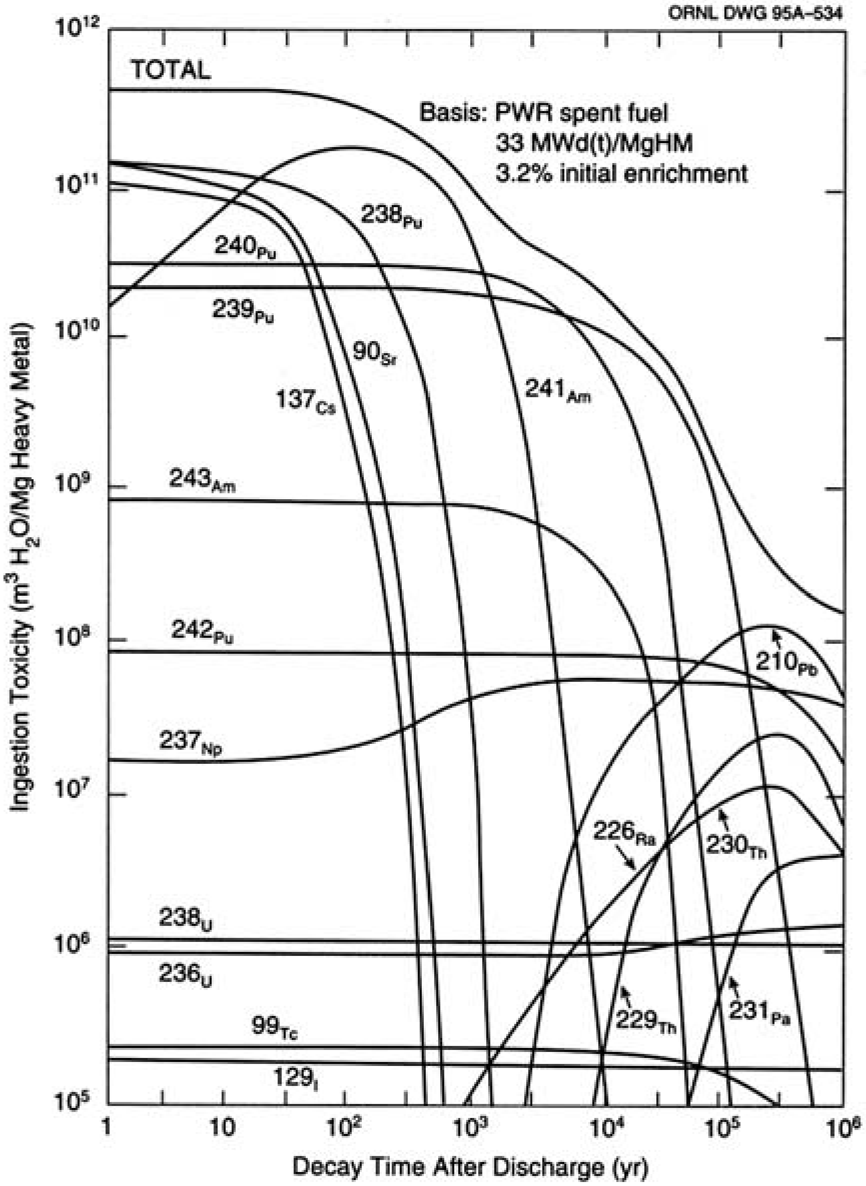

FIGURE 2-2 Toxicity from ingestion as a function of decay time for a number of nuclides in spent LWR fuel. SOURCE: Oak Ridge National Laboratory (1995).

The repository health risk posed by potential ^{99}Tc and ^{129}I releases could be reduced in two ways. First, these radionuclides could be transmuted to innocuous species. This would involve reprocessing spent LWR fuel to recover the radioelements containing these nuclides in relatively pure form, fabricating them into targets, irradiating the targets to destroy part of the radionuclides, and recovering the untransmuted radioelements and recycling them in new targets. The second method of reducing potential releases of these radionuclides is to incorporate them in a waste form specifically tailored to retain them, which would then be

TABLE 2-1 Properties of Selected Fission Products of Interest in Spent Fuel and HLW

Nuclide	Half-Life (years)	Atoms per Fission Product Pair Produced	Thermal Cross Section (barns)
^{99}Tc	2.12×10^5	5.77×10^{-2}	44.4
^{129}I	1.7×10^7	1.07×10^{-2}	37.4
^{90}Sr	28.1	4.43×10^{-2}	1.34
^{137}Cs	30.0	6.02×10^{-2}	0.176

sent to the repository for disposal. This would involve reprocessing spent LWR fuel to recover the radionuclides in relatively pure form, combining them with the appropriate materials to yield a high-integrity waste form, packaging the waste form in a suitable container, and disposing of the waste.

Both options would require development of processes to recover the radioelements Tc and I during reprocessing operations. In addition, the transmutation option would require development and qualification of transmutation targets as well as processes for recycling the untransmuted radioelement. The tailored waste form option for Tc and I would require development and qualification of a suitable waste form and package for the repository. Both of these pose significant but probably not insurmountable challenges.

TABLE 2-2 Properties of Actinides in Discharged Uranium Fuel [a]

Radionuclide	Half-Life	kg	Ci	Elemental Boiling Temperature, °C[b]
^{234}U	2.47×10^{5} yr	3.14	1.94×10^{1}	
^{235}U	7.1×10^{8} yr	2.15×10^{2}	4.61×10^{-1}	
^{236}U	2.39×10^{7} yr	1.14×10^{2}	7.22	
^{237}U	6.75 days	9.15×10^{-7}	7.47×10^{1}	
^{238}U	4.51×10^{9} yr	2.57×10^{4}	8.56	
Total		2.60×10^{4}	α 3.56×10^{1} β 7.47×10^{1}	4,135
^{237}Np	2.14×10^{6} yr	2.04×10^{1}	1.44×10^{1}	
^{239}Np	2.35 days	2.05×10^{-6}	4.78×10^{2}	
Total		2.04×10^{1}	α 1.44×10^{1} β 4.78×10^{2}	—
^{236}Pu	2.85 yr	2.51×10^{-4}	1.34×10^{2}	
^{238}Pu	86 yr	5.99	1.01×10^{5}	
^{239}Pu	24,400 yr	1.44×10^{2}	8.82×10^{3}	
^{240}Pu	6,580 yr	5.91×10^{1}	1.30×10^{4}	
^{241}Pu	13.2 yr	2.77×10^{1}	2.81×10^{6}	
^{242}Pu	3.79×10^{5} yr	9.65	3.76×10^{1}	
Total		2.46×10^{2}	α 1.23×10^{5} β 2.81×10^{6}	3,508
^{241}Am	458 yr	1.32	4.53×10^{3}	
^{242m}Am	141 yr	1.19×10^{-2} yr	1.16×10^{2}	
^{243}Am	7,950 yr	2.48	4.77×10^{2}	
Total		3.81	α 5.01×10^{3} β 1.16×10^{2}	2,880
^{242}Cm	163 days	1.33×10^{-1}	4.40×10^{5}	
^{243}Cm	32 yr	1.96×10^{-3}	9.03×10^{1}	
^{244}Cm	17.6 yr	9.11×10^{-1}	7.38×10^{4}	
^{245}Cm	9,300 yr	5.54×10^{-2}	9.79	
^{246}Cm	5,500 yr	6.23×10^{-3}	1.92	
Total		1.11	α 5.14×10^{5}	—
TOTAL		2.63×10^{4}	α 6.42×10^{5} β 2.81×10^{6}	

[a] Uranium-fueled 1,000-MWe PWR, 150 days after discharge.

[b] G.V. Samsonov [S1].

SOURCE: Benedict et al. (1981).

The Actinides. The important actinides produced in a uranium-fueled reactor are uranium, plutonium, neptunium, americium, and curium. Their half-lives and a calculation of the quantities produced per year in a typical 1,000 MWe PWR are given in Table 2-2. The actinides have a high toxicity index because they are bone seekers when ingested; furthermore, once fixed in the bones, they have a long biological half-life. They irradiate both the bone and the blood-forming cells in the bone marrow, where they may cause both bone cancer and leukemia.

The ingestion toxicity of the actinides of principal concern as a function of time is given in Figure 2-2. For comparison, the ingestion toxicity of the fission products is also shown. As can be seen, the long-term toxicity of spent fuel is dominated by the actinides such as ^{237}Np, ^{234,236}U, and

^{239,240,242}Pu, and the shorter-lived isotopes of thorium, protactinium, radium, lead, etc. that are their progeny. However, the long-term risk is dominated in most scenarios by ^{129}I and ^{99}Tc because, as noted earlier, they are typically soluble and mobile in groundwater pathways. By being relatively insoluble under most conditions, the actinides are not mobile, so despite their high toxicity they contribute very little to the long-term exposure risk in scenarios where groundwater transport is important. There is an exception to this generalization as discussed in Appendix G: the solubility of the actinide neptunium is sensitive to the oxidizing potential as well as pH. Depending on which site properties are used and how infiltration is treated with respect to the availability of oxygen, ^{237}Np, in fact, may dominate the dose in the time frame of around a million years.

In some scenarios, major failure of the repository is postulated. Such failures might be caused by volcanoes, seismic events, or human intrusion, for example. In major-failure scenarios, the actinides might be released by physical damage to the container and the spent fuel. They could then be important contributors to what could be rather high doses to the maximally exposed individual if they are still alive after the more catastrophic events such as a volcano. In such cases, transmutation, which reduces the amount of actinides in the repository, would provide some benefits.

All the actinides will fission in fast-neutron spectrum. A few will also fission in a thermal-neutron spectrum. These latter nuclides include ^{235}U, ^{239}Pu, and ^{241}Pu. Some of the nuclides that do not fission in thermal spectra (e.g., ^{238}U) can be converted into a fissile nuclide by the process:

$$^{238}\mathrm{U} + {}^{1}\mathrm{n} \rightarrow {}^{239}\mathrm{U} \rightarrow {}^{239}\mathrm{Np} \rightarrow {}^{239}\mathrm{Pu}.$$

Nuclides that have this conversion potential property are called fertile. Either a fast or a thermal spectrum can be used to transmute actinides. Because the relative rates of various nuclear reactions differ, the radioactive species in each of the process streams will be different. The amount and type of actinides going to the repository in each case will thus differ, as will the reduction of risk.

Carbon-14. ^{14}C is produced in the atmosphere as a result of a (n,p) reaction on ^{14}N by cosmic-ray-produced neutrons. The total global inventory is about 250 million curies (National Council on Radiation Protection and Measurements, 1985). About 4 million curies of the inventory is in the earth's atmosphere and the atmosphere's annual production rate is about 38,000 curies. ^{14}C is a beta emitter with a half-life of 5,700 years. The global carbon contributes about 1.25 mrem of the natural background dose of about 300 mrem/yr, which includes naturally occurring ^{222}Rn (200 mrem/yr).

^{14}C is produced in nuclear reactors primarily by the (n,p) reaction on ^{14}C, which is present in reactor fuel and structural parts of the reactor as a contaminant. Significant amounts are also produced by (n, α) reactions on ^{17}O that is present in the fuel and by ternary (three-particle) fission. The spent-fuel elements from the once-through fuel cycle to be sent to the repository contain about 1.5 Ci of ^{14} C per Mg of heavy metal. This means that the inventory of 63,000 MTHM of spent fuel would contain about 95,000 Ci of ^{14}C. Calculations indicate that ^{14}C is the major contributor to the dose from gaseous release in an unsaturated repository like Yucca Mountain. When changes in barometric pressure and/or solar heating cause air to move through the repository, any ^{14}C that has been released may be swept out. Models that predict the movement of ^{14}C from the Yucca Mountain repository find that in a few scenarios the proposed EPA limits for ^{14}C are exceeded. It should be noted that the acceptable risk from ^{14}C is not currently set at a level consistent with other standards. This question is currently under review by a National Research Council committee that has responsibilities to recommend to the EPA the technical bases for HLW and spent-fuel disposal standards for the Yucca Mountain repository.

Nonetheless, it seems clear risk to public health from this release of ^{14}C is small. Even if all the repository's ^{14}C inventory (95,000 Ci) were released, the impact would be relatively small. The global airborne inventory would be increased from 4 to 4.095 MCi, which would raise the average dose per person from 1.25 to 1.28 mrem/yr. (The increase in dose would actually be even smaller than given since only a portion of the total release would remain in the atmosphere.) Some people may receive more than the average dose. However, since the repository release would be in the form of CO_2, which is known to rapidly mix in the atmosphere, the maximal dose would still be small. (Maximal and average will both be small, but maximum could be significantly more than average.)

Carbon-14 is essentially untransmutable because its neutron cross section is exceedingly small (about 1 microbarn). If it were to be recovered during spent fuel reprocessing operations, the only possible recourse would be to immobilize it in a specially tailored waste form for repository disposal. However, current practice is to allow it to be released to the atmosphere where, as noted immediately above, it results in a very small dose increase.

The Uranium Problem

A problem that has not received as much attention as it should is the long-term toxicity of mined uranium that is not in the spent fuel. This uranium occurs in two primary materials. The first is uranium mining tailings. Uranium ore typically contains 0.15% to 0.2% uranium mixed with radioactively inert soil and rock. After processing 5% to 10% of the uranium and virtually all of its progeny (e.g.,

^{230}Th, ^{226}Ra) and soil/rock appear in the solid waste from the ore processing facility, which is called tailings. These are typically piled into large mounds which are then stabilized and protected by impermeable caps to retard the release of radioactive radon gas until it decays to a non-volatile radionuclide and prevent the infiltration of water, and large rocks to prevent inadvertent intrusion. If not treated in this way, they can present a major source of public exposure. However, the activity of the tailings decays away with the half-life of ^{230}Th, which is 75,400 years (Croff et al., 1979), and the tailings activity will only decline by a factor of 10 to 20 before the progeny of the very-long-lived uranium isotopes dominate.

The second uranium-bearing material requiring additional attention is tails from uranium enrichment tails. Natural uranium contains about 0.7% ^{235}U whereas LWRs require uranium containing 3-4% ^{235}U. The required ^{235}U concentration is achieved through the uranium enrichment process. However, an inevitable legacy of this process (based on simple mass-balance considerations) is that about 5 kg of depleted uranium remain for each kilogram of enriched uranium product. The ^{235}U concentration in depleted uranium is typically 0.2-0.3%. The United States currently has about 500,000 Mg of depleted uranium stored as UF_6 in metal containers above ground, with most being outdoors. Production continues at a significant rate. If these containers lose their integrity they pose a health risk because contact of UF_6 with water results in the release of toxic fluorine-bearing compounds. Over the very long times characteristic of uranium half-lives the decay progeny will increase to their previous equilibrium levels, although the concentrations will be about 500 times greater than in the parent ore because of the high uranium concentration. The Defense Nuclear Facilities Safety Board (DNFSB) recently expressed concern over the condition and disposition of the enrichment plant tails. Some of the tails have been used for munitions and radiation shielding, and it is proposed that additional amounts be used for multipurpose waste canisters for spent fuel and to provide fuel for advanced breeder reactors. However, it is clear that the existing inventory plus the continued production rate will far outstrip any likely demand for these materials.

The rate at which mine/mill tailings and enrichment plant tails are generated can be reduced by about 33% if the uranium and plutonium in spent fuel are recovered by reprocessing and recycled back to LWRs. The generation rate of mine/mill tails can be reduced to very low levels and enrichment plant tails to zero by the use of advanced breeder reactors that convert the uranium to fissile material on a self-sustaining basis. however, none of the proposed transmutation systems have any plans to deal with the legacy of enrichment tails, and it does not appear possible for them to do so.

SUMMARY OF S&T PROPOSALS AND TECHNOLOGY STATEMENTS

Light-Water Reactor (LWR)

Of all the concepts investigated, one often overlooked alternative is to use existing or advanced LWRs to provide the neutron source to transmute transuranics (TRUs) as well as selected fission products. An LWR TRU-burner could build on the operational experience with plutonium recycle in commercial LWRs in the United States and especially in other countries. Such transmutation potential could be achieved with the least research and development and be available sooner than any of the other concepts considered. Current LWRs could recycle TRUs with mixed-oxide (MOX) fuel in about a third or more of the reactor cores and eliminate significant quantities of TRUs from the existing spent LWR fuel inventories. LWRs could also transmute existing plutonium inventories from the weapons program.

While the fractional transmutation rates in an LWR are not as high as those proposed for the accelerator transmutation of waste (ATW) concept, current LWRs could eliminate TRUs at a significant rate (comparable to an advanced liquid-metal reactor [ALMR] at 0.65 breeding ratio) and still perform within demonstrated engineering practices for materials interactions and thermal power densities. The LWR thermal neutron spectrum can burn certain long-lived fission products, e.g., technetium, that tend to dominate the radiological hazards of a geologic repository. In burning TRUs, however, thermal reactors tend to build up ^{240}Pu, ^{241}Pu, ^{242}Pu, and higher-mass actinides, such as americium and curium. These affect reactivity in the reactor core and are a complication for reprocessing and recycle fuel fabrication and quality assurance. With full recycle of TRUs, the amounts of ^{240}Pu and ^{241}Am could significantly reduce reactivity. Any reactivity deficiency could be remedied by mixing the recycle TRUs with uranium of moderate enrichment, which would also enhance the capability for burning the long-lived fission products.

An LWR transmutation system could use the most well-known approach to reprocessing based on aqueous separations technology. A new generation of reprocessing plants could be applied to existing spent LWR fuel and could support subsequent application of more advanced transmutation concepts. Also, the reprocessing would allow development of optimized low-water leachable waste forms to enhance the safety of the disposal of untransmuted nuclear waste in a geologic repository.

Advanced Liquid-Metal Reactor (ALMR)

The advanced liquid-metal reactor/integral fast reactor (ALMR/IFR) program had been sponsored by the Depart ment of Energy/Nuclear Energy (DOE/NE) during the previous administration and was under joint development by GE and ANL. In 1994 the ALMR/IFR program was cancelled. Limited funds were appropriated for phase out and continuation of research on separations.

The ALMR has been under development for decades as a breeder reactor, but it could be modified to operate as an efficient TRU burner. Its net consumption rate increases as its breeding ratio is reduced from the usual values near unity. At a breeding ratio of 0.65, the ALMR would be comparable to an LWR in net TRU consumption rate. The ALMR, as part of any IFR system, includes pyroprocessing of its spent fuel. This technology has not yet been demonstrated at pilot scale. the possibility of success is intermediate, that is, less than that of the LWR but greater than that of the ATW concepts. An ALMR/IFR system prototype would also require an intermediate time and cost to complete development and full-scale demonstration.

The ALMR fast-neutron spectrum has both advantages and disadvantages for transmutation, compared to the thermal spectrum of an LWR or ATW. The ALMR would produce much less ^{240}Pu, americium, and higher-mass actinides. However, the ALMR would have impractically low rates of transmutation for long-lived fission products, including those that dominate the radiological hazard with a geologic repository. The ALMR presents safety issues peculiar to a fast, liquid metal-cooled critical reactor, in particular, the issue of the sodium void coefficient. The predicted performance appears acceptable for transmutation at breeding ratios near unity. However, considerable experimental and calculational effort at breeding ratios of 0.65 or lower would be necessary to develop a database to support licensing with a reactor core that is fueled for full recycle, as proposed by the ALMR/IFR program, containing higher-mass TRUs and fission products such as the lanthanides.

The choice of an integrated all-pyroprocessing (see Chapter 3 and Appendix D for full details of pyroprocessing and other separation processes) system for an ALMR/IFR is quite a departure from the previously well-studied approach with aqueous separations technology. Pyroprocessing has been used extensively with metallic fuels in the DOE weapons programs and has been demonstrated at laboratory scale for ALMR metallic fuel. However, cost-effective pyroprocessing has not been fully developed for mixed-oxide (MOX) LWR spent fuel, which has a TRU content of only about 1% heavy metal (HM) compared to about 30% for the ALMR metallic fuel. Pyroprocessing requires considerable further development and scale-up with full pilot-plant demonstration, especially for spent LWR fuel application to commercial processing. As a low threshold cost for LWR spent-fuel reprocessing is required for a viable ALMR/IFR burner system, its probability of success as a practical, economic system is lower than that of an ALMR for power production. Indeed, the latter could be started from existing stores of plutonium (or even from highly enriched uranium [HEU]) without reprocessing spent LWR fuel.

Accelerator Transmutation of Nuclear Waste (ATW)

The ATW proposals project exceptionally high fractional rates for transmuting TRUs and long-lived fission products. The proposals aim at reducing the TRUs in waste to such an extent that all waste containing residual TRUs would be Class C or less than Class C, i.e., suitable for shallow land burial. However, the transmutation rates require extraordinary operating conditions in the ATW subcritical reactor and beam target (e.g., a factor of 10 higher thermal neutron flux and 2 1/2 higher power density than in a commercial LWR), posing major engineering and materials challenges. In addition, the ATW would use fluid fuels, an unproven technology that presents problems of reprocessing and reconstituting the fuel and raises operational and safety issues. The on-line reprocessing system has unique requirements for reliability and maintainability, but a detailed concept does not yet exist. Thus, extensive research and development would be required even to ascertain whether an ATW is feasible. Compared to an LWR or ALMR, the ATW concepts have a much lower chance of success as a practical, economic system and would require the longest time and highest cost to complete development and full-scale demonstration.

Major development issues confront the fuel system and structural materials. The ATW concepts differ primarily in the type of fluid fuel employed. In one ATW concept, the fuel is a slurry of tiny waste particles in heavy water circulating in small tubes under high pressure. Nonhomogeneity in the slurry and erosion of tube walls caused the failure of attempts to use this type of approach decades ago in Europe. Also, the ATW would produce more higher actinides than an LWR; the high alpha and neutron activity would degrade the aqueous fluid and reagents and greatly complicate recycle fuel qualification. The more advanced ATW concepts circulate a molten salt with dissolved radionuclides, which would be more radiation resistant. However, this approach suffered corrosion problems in reactors at Oak Ridge National Laboratory (ORNL) in the 1960s and 1970s. Moreover, the structural materials must withstand conditions of neutron flux and fluence beyond engineering experience (e.g., more than 100 displacements per atom) that degrades basic properties and imparts dimensional instabilities.

Intense beams of high-energy protons from an accelerator would be focused on targets such as lithium, tungsten, or molten lead, which would generate high-energy neutrons. These would be thermalized and multiplied in a subscritical reactor to transmute radionuclides. The exceptionally high volumetric fission rate entails high-power densities in the reactor in addition to high thermal power in the beam target. Thus, heat removal is a major development and safety issue. In addition, the possibility of reactivity transients in the ATW may call for reactivity control measures beyond merely shutting off the accelerator as explained in Chapter 4 and Chapter 6. Overall, the safety-related problems are severe enough that licensability of an ATW seems doubtful under present regulatory practices.

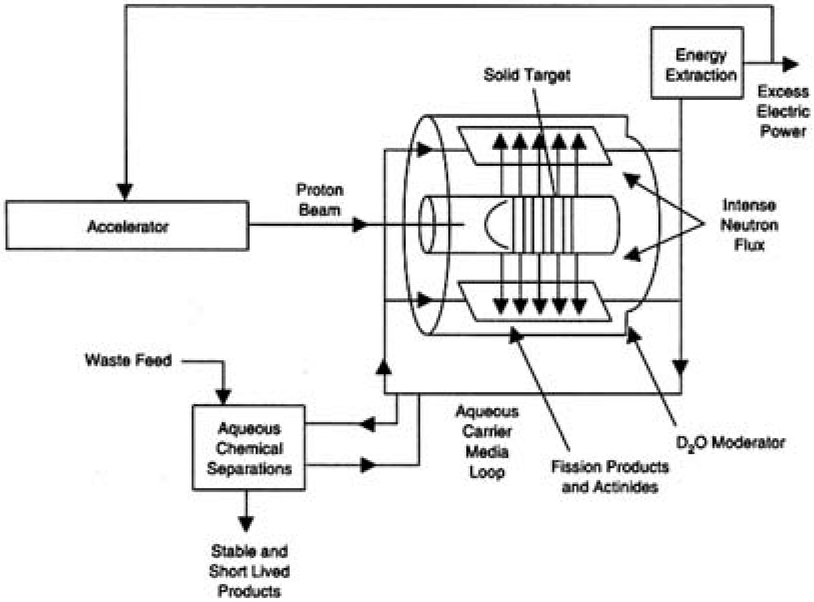

FIGURE 2-3 Reference design for commercial waste transmuter.

It should be noted that the LWR transmutation system envisions using essentially current LWRs with some changes in the fuel composition. As in all transmutation systems, reprocessing would be required. The ALMR system would follow an ANL conceptual design that would employ metal fuel and pyroprocessing. A commercial system like this has never been built and operated; however, experience with similar systems gives confidence that such a system could be designed and built. The ATW is so drastically different in design that there is essentially no experience with such a system. Figure 2-3 shows a schematic of one of the ATW systems. The accelerator is a 1.6-MeV proton machine with a beam current of 0.25 amp. The protons hit a heavy target and produce neutrons. The neutrons are then multiplied by a factor of 20 by a subscritical assembly. The energy dissipated in the target is about 8,000 Mw. The heat is recovered to produce the power for the accelerator, and the excess power is sold to the grid. The material to be transmuted is an aqueous slurry that is processed and fed back into the system. If the designed flux of over 10^{15} n/cm^2-s is achieved, this system would be able to attain much higher burnup rates than either of the other systems (see Appendix F for details).

Separations Technology and Commercial Spent-Fuel Reprocessing

At present, there are the only four industrially demonstrated separations processes applicable either to DOE defense reactor wastes or to meeting the needs of transmutation of TRUs and fission products in spent LWR fuel. The main steps in reprocessing spent LWR fuel are shown in Figure 2-4. These processes are designed primarily for the concentration and purification of plutonium, but only the PUREX process is well established in current worldwide use. In the past, the British have used a solvent extraction process called BUTEX, the French have used ion exchange, and there have been a number of ion exchange processes that have had limited production use in the isolation of minor actinides.

Several potentially applicable separations processes based on new solvents, such as the TRUEX-CMPO (carbonmayl methyl phospine oxide) process, and new ion-exchange materials are in various conceptual or laboratory-scale developmental stages. Such advanced aqueous processes have been proposed to achieve high decontamination factors for TRUs but have not been demonstrated at the full engineering pilot-plant level. Moreover, aqueous technologies would need further development to cope better with the radiation environment accompanying the higher-mass actinides. Among advanced nonaqueous technologies, pyroprocessing has been used extensively in the DOE weap ons programs with metallic fuels (pyroprocessing is described in detail in Chapter 3 and Appendix E). Argonne National Laboratory (ANL) has a pyroprocessing development effort for ALMR spent-fuel reprocessing under way; an extension of that process is being studied by ANL for possible employment in the reprocessing of LWR spent fuel.

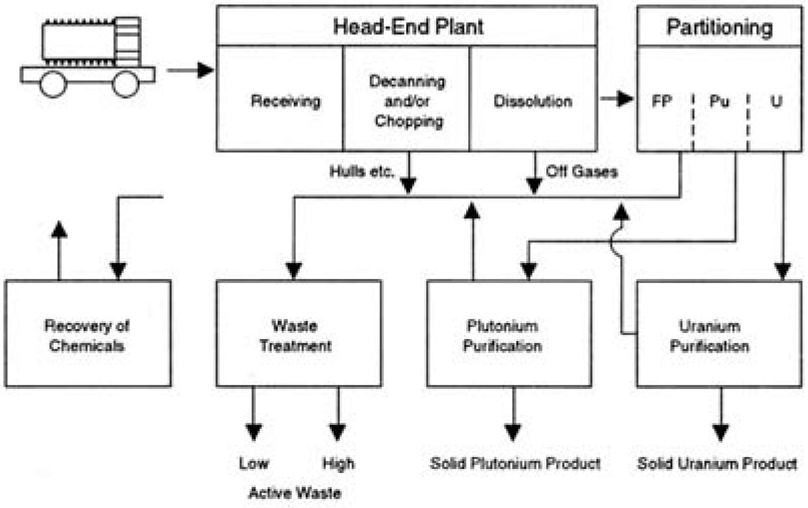

FIGURE 2-4 Main steps in reprocessing spent LWR fuels.

The transmutation concepts being proposed require headend treatment of spent LWR fuel to chop and dissolve the fuel, followed by separation of the TRUs and selected fission products. Either aqueous or nonaqueous processes may be used for the initial separations. The well-established PUREX process may be useful for this separations step. For the initial processing of recycle oxide fuels, even PUREX needs further development to overcome difficulties in dissolving MOX.

For the LWR transuranic burner system, an initial PUREX separations process could be followed by an aqueous separations process using an advanced technology, such as a TRUEX-like process, to achieve high decontamination factors for TRUs. An additional specialized stage would be needed to separate technetium, if desired. Development and pilot-scale testing of the latter two processes, plus demonstration of an overall separations system at engineering pilot scale, would be necessary before a full-scale separations system could be designed with high confidence, especially to meet stringent goals for very low overall separations process losses (0.1 % or smaller).

For the metallic fuel used in the ALMR, a number of pyrochemical separations for reactor spent-fuel processing have been demonstrated to a several-kilogram-batch scale with mock fuels. These methods have worked essentially as the basic thermodynamics predicts, with many exhibiting very high separation factors. However, as yet, there has not been extensive development of the equipment and techniques for the multistage cascades needed for the high separation factors entailed in the DOE ALMR/IFR proposal. In addition, ANL proposes pyrochemical processing of spent LWR fuel. The process for the LWR fuel application is less mature than for the metallic ALMR fuel and requires extensive testing, scale-up, and demonstration to achieve a viable ALMR/IFR system.

The separations processes required for the ATW transmutation concept are substantially more difficult than those required for other proposed transmutation concepts (i.e., the ALMR and LWR). This is due to the much more intense radiation fields in the ATW, which destroy many reagents involved, and the low process losses required under these conditions to meet the goals of the ATW proposals. The aqueous concepts that have been suggested are so far beyond demonstrated process experience in such radiation fields as to be highly questionable. In addition, the ATW reprocessing system requires a mechanical chopping subsystem and an industrial-scale TRUEX-like separations process to obtain the TRUs from spent LWR fuel for start-up and make-up.

In addition to separations facilities for spent reactor fuel, recovery plants will have to process the miscellaneous salvage materials generated from the processing of work material, the repair of equipment, the impurities in reagents, and the corrosion products. The recovery processes will be expensive to carry out, and those needed will not be well known until there has been extensive pilot-plant testing. A very high degree of recovery of active elements from these salvage materials will be necessary to meet cycle recoveries needed for low overall process losses.

An additional technical uncertainty arises from the lack of regulatory experience with separations plants. Environmental and safety requirements necessary to meet concerns identified in the licensing and regulatory process could impose unknown technical obstacles and burdens on design and operation of separations plants as a part of a total separations and transmutation system.

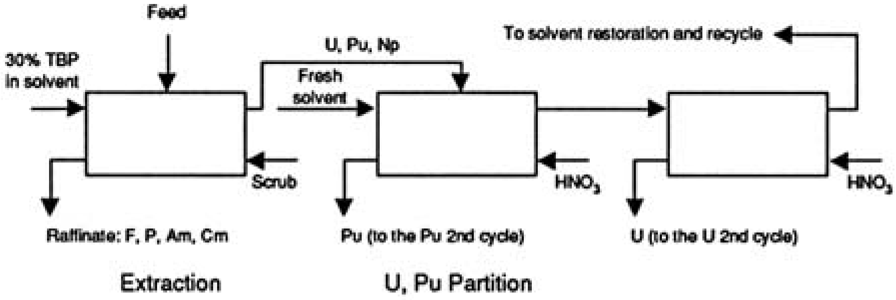

FIGURE 2-5 Flowsheet for the first purification cycle in the PUREX process. SOURCE: Musikas et al., 1992.

Separations and Transmutation of Plutonium Only

Either an LWR or an ALMR would operate much more efficiently with ^{239}Pu as the recycle fuel than with recycle of all the TRUs contained in the LWR spent fuel. This suggests a waste management concept intermediate between the once-through LWR fuel cycle and the full transmutation of all TRUs and selected fission products, i.e., the accumulated LWR spent fuel could be reprocessed and only the separated plutonium recycled to either an LWR or ALMR. A flowsheet for the first purification cycle in the PUREX process is given in Figure 2-5. The recovered uranium would be packaged for disposal or possibly for recycle to an enrichment facility. In addition, ^{14}C and ^{129}I could be captured, e.g., as off-gasses, and packaged in low-solubility waste forms that could be developed. Such an LWR could also transmute the long-lived ^{99}Tc and ^{129}I fission products if separated and recycled. Of course, the big cost is that of spent LWR fuel reprocessing and the all-remote fabrication of plutonium-bearing recycle fuel. Moreover, the concept opens the door to major public policy issues entailed with the commercial use of plutonium, in particular, its relationship to U.S. nonproliferation policy.

For such plutonium-only transmutation, present-day separations technology using PUREX may be sufficient, with process losses of the order of 1% for plutonium.[2] Some of the neptunium might also be recycled if that proved easy to do. The remaining TRUs and the bulk of the fission and activation products would go to high-level radioactive wastes (HLW); this PUREX waste might be contained in a glass waste form as planned for the defense HLW. By also separating certain long-lived fission products, one could possibly devise other improved waste forms, such as for ^{99}Tc and ^{135}Cs. If pyroprocessing were used with an ALMR plutonium burner, other waste forms would need to be developed.

It is more problematical whether such plutonium-only transmutation could capture some of the prospective benefits to waste management claimed for the full transmutation of the TRUs. First, transmuting the plutonium could reduce somewhat the calculated repository hazards from human intrusion scenarios in which waste is brought directly to the surface. However, the effect would depend on the time after repository closure at which the human intrusion event occurred. For postclosure times of about a hundred years or so, ^{238}Pu with an 87.7-year half-life tends to dominate the TRU activity and would be transmuted. However, for postclosure times of a few hundred years to several thousand years, the untransmuted ^{241}Am with a 433-year half-life would dominate the TRU activity. This would be especially true for the older spent fuel in which the ^{241}Pu had already decayed with a 14.4-year half-life to ^{241}Am. For postclosure times of ten thousand years or more, ^{239}Pu and ^{240}Pu would dominate the TRU activity and would be transmuted. Over very long periods of time, when the ^{239}Pu is eliminated, ^{237}Np would dominate the transuranic activity.

In addition, under planned repository conditions, plutonium is quite insoluble in groundwater. Thus, plutonium transmutation would have little direct effect on the important long-term repository risk scenarios in which groundwater dissolves radionuclides and transports them to the biosphere, except by eliminating one of the precursors of ^{231}Pa. Also, removal of the plutonium would reduce the long-term heat load on the repository, although the untransmuted ^{241}Am would still be a major contributor. For the Yucca Mountain repository, however, decay heat is being considered as a means to keep the waste containers dry while increasing the capacity of the repository, so that transmuting the plutonium (or other TRUs) may actually be a disadvantage.

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[2] The National Academy of Sciences had another study on weapons plutonium, and a report entitled, "Management and Disposition of Excess Weapons Plutonium" was published in 1994.

Defense High-Level Waste

Practical separation processes for DOE defense wastes stored in the Hanford tanks appear to be available. The Hanford tank wastes exist as mixtures of aqueous solutions, salt cakes, and sludges. Water washing of the sludge and dissolution of the salt cake will remove most of the ^{137}Cs and some of the ^{90}Sr (the principal radioactive contaminants) from the tanks, facilitating their separation and disposal as HLW and leading to a large reduction in HLW volume.

Additional processing of tank wastes, beyond sludge washing, requires dissolution of the sludge in acid. Subsequent separation of radioactive, and perhaps other, contaminants appears feasible, using processes not yet fully developed or demonstrated. A program of further research and development, ranging from laboratory studies for some proposed separations to pilot-plant development for others, is needed before design of a final separations system. It is expected that there will be appreciable insoluble residues left in some of the tanks after sludge washing. Attempts to remove these residues could seriously damage the tanks.

RADIONUCLIDES AND REPOSITORY PERFORMANCE

LWR Spent Fuel Characterization

The base case for the evaluation of the proposed transmuter systems is the once-through uranium fuel cycle for the LWR, with geologic disposal of the spent fuel. After a few years, the decay of the radionuclides in the spent fuel and the associated heat load on the repository are dominated by two fission products, ^{137}Cs and ^{90}Sr, with half-lives of about 30 years each. For example, in 10-year-old spent pressurized water reactor (PWR) fuel with a nominal burn-up of 33,000 MW-days/MgU at discharge, the ^{137}Cs and ^{90}Sr decays produce about 90 w/MgU and 65 w/MgU, respectively[3] (Rawlins et al., 1990). After about 75 years, the heat load begins to be dominated by decay of TRUs, primarily several plutonium isotopes and the americium component of the minor actinides (MA).[4] For the reference 10-year-old spent PWR fuel, Table 2-3 lists five TRUs that are important for heat production. In a repository the thermal power integrated from 10 to 1,000 years comes mainly from the following sources: ^{241}Am (including the parent ^{241} Pu), 44%; ^{137}Cs and ^{90}Sr, 21%; ^{240}Pu, 10%; ^{238}Pu, 7%; and ^{239}Pu, 7%. After 1,000 years, the relative contributions from ^{240}Pu and ^{239}Pu would continue to grow while those from the other sources diminish.

TABLE 2-3 Heat Release from Selected TRUs in Spent PWR Fuel (33,000 MW-days/MgU exposure—10-year decay)

Isotope	Half-Life (years)	Heat Release (W/MgU)
^{238}Pu	87.7	78.0
^{239}Pu	2.411×10^4	9.7
^{240}Pu	6,560	16.5
^{241}Pu	14.4	2.46[a]
^{241}Am	433	57.3

[a] ^{241}Pu is an important source of ^{241}Am by beta decay.
SOURCE: Rawlins et al. (1990).

The maximum attainable temperature of a geologic repository depends on the heat capacity and thermal conductivity of the geologic medium; the age, amount, and density of the spent-fuel loading; and the ventilation practices during operations. For a representative spent-fuel loading planned for the unsaturated tuff at Yucca Mountain, the maximum temperature at the repository center would occur after about 1,000 years and would remain above the boiling point of water for about 2,000 years (Ramspott et al., 1992). If the TRUs were removed, the integrated thermal power from 10 to 1,000 years would decrease by more than a factor of 4. The corresponding repository center temperature would reach the boiling point of water for a brief time and drop below 50°C within a few hundred years (Ramspott et al., 1992).

DOE currently plans to package spent LWR fuel rods in sealed containers for placement in the repository. To satisfy code of NRC regulations 10CFR60, the containers must have high integrity during emplacement and for the decades or so that the repository is being filled, such that the containers could be removed safely if the repository were found to be defective. The NRC regulations also address the integrity of the waste package after the repository is permanently closed and sealed. These regulations require that "… the engineered barrier system shall be designed, assuming anticipated processes and events, so that (A) Containment of HLW within the waste package will be substantially complete for a period to be determined by the Commission and that period must be in the 300 to 1,000-year range after permanent closure of the geologic repository …" (10CFR60.113). The Yucca Mountain program has set a performance goal of 1,000-year integrity for the spent-fuel waste package. From a regulatory point of view, however,

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[3] The units for isotope concentrations and heat release are normalized to initial megagrams of uranium in the fuel.

[4] The minor actinides considered here are neptunium and protactinium plus the elements with an atomic number greater than that of plutonium—especially americium, curium, berkelium, and californium.

it may not be easy to prove package integrity in a repository for much longer than a few hundred years after closure.[5]

Estimating Radiological Repository Risk

The health and safety risks associated with HLW in a geologic repository are discussed in Chapter 6. The estimates of such risk focus on the increased radiation dose to which people might be subjected in the future under various scenarios in which radionuclides might escape the repository and reach the accessible biosphere. Such a scenario analysis, using probabilistic risk assessment, plays an important role both in formulating the generic criteria that all repository sites must meet and in evaluating a particular site, such as Yucca Mountain.

Radionuclide Release Scenarios

Radionuclide release scenarios take into account a wide range of possible events. The two time scales for the repository risk, using the 10,000-year EPA time horizon as a divider are:

- extending for a period of 10,000 year after closure,
- extending beyond 10,000 years to hundreds of thousands of years.

The half-lives and main sources of various TRUs are listed in Appendix G, Table G-2. For either time period, the three types of radionuclides of importance for repository risk are:

- *plutonium*, i.e., ^{238}Pu, ^{239}Pu (which is the principal fissile component in the spent fuel), ^{240}Pu, and ^{242}Pu;
- *minor actinides*, i.e., ^{237}Np, ^{241}Am, and ^{243}Am, plus even higher-mass minor actinides in some transmutation schemes; and
- *fission products*, mainly ^{137}Cs and ^{90}Sr for a few hundred years, plus long-lived soluble fission products, especially ^{99}Tc (2.1×10^5 yr) and ^{129}I (1.6×10^7 yr) and, to a lesser extent, ^{135}Cs (2.9×10^6 yr).[6]

Of the scenarios relevant to the long-term repository risk, the most important involve the dissolution of the HLW in groundwater and the transport of the dissolved radionuclides to the accessible biosphere, so-called *dissolution-and-migration* scenarios. Such events could arise not only with geologic media that are normally saturated with groundwater, but also with initially dry media, if, for example, it is infiltrated with water or when there are long-term changes in the water table. For this class of scenarios, risk estimates show that the isotopes of primary concern are two long-lived, soluble fission products, ^{99}Tc and ^{129}I (Croff et al., 1980). To a lesser extent, long-lived ^{135}Cs could also contribute, but its groundwater transport would be retarded by an estimated factor of 360 due to sorption on rock during the migration (Pigford, 1990).

The TRUs have significant radioactivity during this period, especially the long-lived minor actinide ^{237}Np (2.14×10^6 yr half-life). If reducing conditions exist in a repository the transuranics tend to be relatively insoluble in groundwater and would not be transported to the biosphere in significant quantities.[7] For an unsaturated repository with oxidizing conditions, ^{237}Np may be an order of magnitude more soluble than assumed in earlier analysis based on reducing conditions (see Appendix G for details).

Calculations for a granite repository, discussed in Chapter 6 (see section *Impact of S&T on Waste Repository*) and Appendix G, indicate that the main actinide contributor is not ^{237}Np but rather ^{231}Pa, which grows in spent fuel from the decay of its precursors ^{235}U, ^{239}Pu, and ^{241}Am. For the Yucca Mountain site, there is no reason to think or assume that ^{231}Pa would be any less important for tuff than for granite, given the different conditions of the formations. Thus, ^{231}Pa is likely to be the radionuclide of significance for the dissolution-and-migration scenarios, if the long-lived fission products were greatly reduced.

For these reasons, the various transmutation concepts have been evaluated for the conditions under which they could achieve a reduction of several orders of magnitude in the amounts of ^{99}Tc and ^{129}I, i.e., down to the range at which the TRUs begin to appreciably affect the estimated long-term repository risk.

The second major class of scenarios is that in which the integrity of the repository is compromised, the so-called *repository-breach* scenarios, which involve bypass of the normal pathways to the biosphere. These scenarios could be caused by, for example, natural events such as earthquakes and volcanism. However, the repository site would be cho

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[5] Sweden is planning a much more elaborate and expensive waste package for the spent fuel, comprising a lead-filled copper container with a wall thickness of 5 to 10 cm, which is highly resistant to corrosion, surrounded by absorbing bentonite clay. The Swedish program anticipates that the integrity of the waste package in a hard-rock repository could be ensured for many thousands, perhaps millions of years (see Chapter 6).

[6] HLW also contains other long-term fission products of less importance, such as ^{79}Se ^{93}Zr, ^{107}Pd, and ^{126}Sn, plus long-lived activation products such as ^{59}Ni, ^{93}Mo, and ^{94}Nb. Also, ^{14}C may be an important activation product for the special case of the Yucca Mountain repository, as discussed in the text (see also Chapter 6 and Appendix G).

[7] Decay daughters from uranium (e.g., ^{226}Ra and ^{231}Pa) are also predicted (Svensk Kärnbränslehantering AB [Swedish Nuclear Fuel and Waste Management Co.], 1992\; Statens Kärnkraftinspektion [Swedish Nuclear Power Inspectorate], 1991) to contribute to long-term doses. Transmutation of the uranium parents is not proposed, but reprocessing to accomplish transmutation would reduce the amount of uranium in the repository by a magnitude of the order of the separations process decontamination factor.

sen to minimize the significance of such natural events (among a variety of other selection criteria). Another potential cause is human intrusion, such as in drilling for water or other extractable resources. Studies are not yet definitive about the significance of human intrusion compared with other release scenarios. One study for a repository in granite indicates that human intrusion scenarios may have significantly less weight in the overall repository risk than dissolution-and-transport scenarios. For example, the risk assessment for Yucca Mountain considers human intrusion scenarios in which radionuclides could be brought to the biosphere in the vicinity of the repository and made accessible over a large area, e.g., by resuspension and wind transport (see Chapter 6 section on *Impact of S&T on Waste Repository* , and Appendix G). In more complex situations, some repository-breach scenarios can connect with dissolution-and-migration scenarios, for example, the breach of a barrier between the repository and a nearby aquifer, which enables radionuclides to reach the aquifer and be readily transported by groundwater.

For human intrusion scenarios the TRUs could be important in the calculation of repository risk, especially plutonium and americium isotopes (as well as ^{237}Np for the long-term). Thus, this chapter assesses the effectiveness of the various transmutation concepts for reducing the inventory of TRUs. However, such a reduction would not simply produce a proportionate reduction in repository risk for repository-breach scenarios. Transmutation can reduce the amounts of some TRU isotopes while increasing the amounts of others. The net result depends on the scenario and the amounts of the various TRU isotopes remaining after transmutation.

Gaseous release of the radionuclide ^{14}C is currently an issue under discussion for the Yucca Mountain repository (see Chapter 6 section on *Impact of S&T on Waste Repository*). ^{14}C is produced by neutron absorption in the ^{14}N contained in the uranium dioxide in spent fuel and in the Zircalloy fuel cladding. Although spent fuel does not have a large curie inventory of ^{14}C, the calculations assume that essentially all of it would be released as a gas during a few thousand years under oxidizing conditions such as in the unsaturated tuff at the Yucca Mountain site. The dose to an individual living on the surface would be small, but under the original standard, the calculated probabilistic cumulative release was considered important. If the spent fuel were reprocessed, the ^{14}C could be captured as an off-gas and packaged in a low-release waste form for geologic disposal.

Regulation 10CFR60, together with the U.S. Environmental Protection Agency (EPA) standard for protection of the accessible environment (40CFR191), also put stringent limits on the release rates and the cumulative releases of the radionuclides present in the repository after the containment period, taken here to be 1,000 years. Until recently, the EPA standard governing a geologic repository required compliance with cumulative release limits for categories of radionuclides. To establish the limits, the EPA considered many specific pathways to the accessible biosphere for each of the three principal types of pathways—gaseous transport, groundwater use, and human intrusion—and estimated the amount of each radionuclide that would have to be released to cause the equivalent of 1,000 premature deaths over a 10,000-year period. Such estimates entail assumptions about future populations and have sparked controversy about the practicality and acceptability of the basis of the EPA standard 40CFR191 (see Chapter 6).

In October 1992, Congress exempted the Yucca Mountain repository from the current EPA standard 40CFR191 and directed the EPA to commission a National Research Council study to provide findings and recommendations concerning the technical basis for health-based standards (The Energy Policy Act of 1992—PL 102-486). The National Research Council study is to consider whether a health-based standard based on doses to individual members of the public will provide a reasonable standard for protection of public health and safety. Congress also directed the Nuclear Regulatory Commission (NRC) to modify its technical criteria and requirements to become consistent with the new EPA standards. For either dose basis, however, one can identify the key radionuclides that most affect the dose estimates. The relative effectiveness in reducing the amounts of these key radionuclides can thus provide a useful measure to compare the performance of the various transmutation concepts. Individual dose and risk standards have already been directly incorporated into repository evaluations in other countries. In all cases, the allowable amount of radioactivity that can be introduced into the environment is small. The original EPA standard was consistent with the goal of 1,000 cancer deaths in 10,000 years from 100,000 metric tons uranium (MTU) of spent fuel or equivalent.

Individual dose standards are usually less than background, and in that sense are a fraction of background. However, they are set at a numerical value rather than at a fraction of background.

An Optimized Waste Form

In an S&T system, a waste form optimized for geologic disposal could be used for the HLW remaining after transmutation. Of course, separations by itself could yield waste forms that could, in principle, encompass a material for each constituent radionuclide that maximizes its chemical and physical integrity in the overall waste package. An ideal optimized waste form after separations would be solubility-limited in groundwater for key radionuclides that affect re pository risk significantly. For example, AgI has low water solubility and could be used as the waste form for separated ^{129}I to greatly reduce its fractional release rate in a repository.

Indeed, a combined reduction-and-packaging strategy could be considered in which some constituents are transmuted and their remaining HLW optimally repackaged for geologic disposal, together with optimal materials chosen for other separated radionuclides that are not transmuted. Such a strategy would greatly increase the number of options open to a system designer, but puts a premium on the ability to predict waste-form behavior for very long time periods. However, as noted in Chapter 6, *Impact of S&T on Waste Repository* , there is no evidence from the assessment to date of repository risk that S&T systems and/or optimized waste forms are actually needed to improve nuclear waste disposal relative to the once-through LWR fuel cycle.

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Chapter Three

Separations Technology

This chapter addresses the role separations technology can play in the management of spent light-water reactor (LWR) fuel and defense wastes to help reduce their long-term risk to humans and their environment. Additional information on aqueous and nonaqueous processes, reprocessing experiences in the United States and abroad, and future separation processes is given in Appendix D of this report.

It may be acceptable to dispose of spent LWR fuel with currently known mechanical disassembly methods and encapsulation (packaging) technology for the foreseeable future. However, for some defense wastes that have arisen as a result of processing of reactor target fuels, it may be necessary to separate a large-volume, low-level radioactive fraction (low-level waste, or LLW) and a much smaller-volume, high-level radioactive fraction (high-level waste, or HLW). The perceived advantage of this approach to managing the wastes is that a smaller volume of the highly radioactive fraction (in a suitable waste form, perhaps as a vitrified glass product) needs to be disposed of in a relatively expensive repository; the larger low-level radioactive fraction can, after appropriate treatment, be safely disposed in a much less costly manner. The overall safety is expected to be increased and the disposal cost minimized by this approach.

If it is decided to prepare the fractions for reuse as targets for transmutation or for discharge to a waste-disposal system, it is necessary to separate the highly radioactive waste materials in them into specific components. In general, separation of transmutation targets would demand a much higher degree of purification than would preparation of repository waste. A number of processes have been proposed for carrying out the needed separations. Some of them have been demonstrated at the bench scale and not necessarily with the levels of radioactivity that would be present in the actual wastes; a few others have been demonstrated on a very large scale at high levels of radioactivity and used on an industrial scale. For example, the plutonium and uranium recovery by extraction (PUREX) process, an aqueous process employing solvent extraction techniques, was developed for, and has been used extensively, in reactor target processing for plutonium production for weapons in the United States and for weapons and commercial fuel recycling in power reactors in France, the United Kingdom, and the former Soviet Union. Pyroprocesses, which typically employ high-temperature, nonaqueous systems, have been used for many years in the nuclear as well as in conventional industries, for example, for electrorefining of plutonium and americium—though they have had limited use for processing of high-level waste requiring remote operations.

In all cases, it is necessary to tailor the processes to the specific separations required. In most of the cases, substantial development and demonstration of the processes on the actual wastes will be necessary, since many of the new process objectives differ from the original process objectives.

HISTORICAL BACKGROUND

The development of large-scale radiochemical separations technology was begun in 1942 by the scientific staff of the Metallurgical Laboratory, a part of the Manhattan Engineering District organization that was created during World War II to develop nuclear weapons for military applications. The mission of the Separations Section of the Metallurgical Laboratory was to develop an industrial-scale chemical process to recover and purify plutonium from the uranium targets that were to be irradiated in the "neutron chain reactors" proposed for construction at Hanford, Washington (Thompson and Seaborg, 1956). The underlying separations principles and many of the techniques devel oped and used at that time are essentially the same as those in use today.

Much of the basic chemistry of thorium and uranium was known in 1942, but the nuclear decay characteristics of most of the fission products were not. Furthermore, the chemistry of many of the fission products and transuranic (TRU) elements was not known in sufficient detail. Promethium, technetium, and all the TRU elements were new to science and much had to be inferred from an element's position in the periodic table. The chemical and physical effects of radiation imposed additional difficulties and uncertainties in the proposed processes, as they do even today.

The concentration of the plutonium to be produced in the reactors at Hanford was quite low, on the order of 300 grams of plutonium per ton of irradiated uranium. (The only plutonium available for process development activities prior to the construction of the graphite reactor in Oak Ridge in 1943 was about a milligram of ^{239}Pu created by extended cyclotron bombardment of uranium.) The separations processes had to have very high recovery efficiencies, while also achieving very high separation of the plutonium from the radioactive fission products that were produced in the uranium targets along with the plutonium. It was a monumental challenge to the Metallurgical Laboratory chemists, most of whom were new to this field of industrial-scale separations chemistry.

Reprocessing technologies can be grouped into two categories: those based primarily on aqueous chemistry and those based on non-aqueous materials such as molten salts and molten metals. A summary description of the major approaches in each category is given below. A detailed description is given in Appendix D.

Aqueous Processes

The chemical differences between the several oxidation states of plutonium have been extremely important in separations. In 1941 it was shown that plutonium, like uranium, has multiple oxidation states (Hill and Cooper, 1958) and coprecipitates with lanthanum fluoride in its lower valence states, but not in its oxidized states. In the IV oxidation state, plutonium was found to coprecipitate with the hard-to-process gelatinous iron hydroxide and zirconium phosphate. In 1942, a process based on bismuth phosphate ($BiPO_4$) precipitation was successfully developed. It was the first large-scale radiochemical separation to exploit oxidation-reduction technology for both purification and the concentration of a single element from many other contaminants. However, the bismuth phosphate process, while successful, recovered only plutonium; the valuable uranium in the irradiated feed became waste. An extraction process based on the ability of the organic solvent methylisobutyl-ketone (hexone) to extract both plutonium and uranium (and neptunium as well) from oxidizing solutions was developed next (Hill and Cooper, 1958); it recovered both uranium and plutonium and had less complicated remote mechanical operations. Because it used oxidation-reduction chemistry, this method was referred to as the REDOX process.

A large number of solvent extraction systems were investigated subsequently (Culler, 1956). Much effort was concentrated on the alkyl phosphates after it was found that tributyl phosphate (TBP) had superior characteristics over other solvents for extracting both plutonium and uranium. TBP therefore became the extractant of choice in the PUREX process, which eventually became used internationally. An advantage of TBP over the methylisobutyl-ketone (hexone) used in the REDOX process was that it allowed the use of nitric acid as the process salting agent. Most of the other extractant systems required aluminum, magnesium, or some other highly soluble nitrate salt in concentrated solution (which would contribute to the waste problem).

Nonaqueous Processes

Nonaqueous processes have been used in reprocessing plants to separate materials for nuclear weapons and in plants for reprocessing spent reactor fuels, mostly to clean up the uranium product. Some of these processes are high-temperature processes (hundreds of degrees centigrade), and they are referred to as either pyrochemical or pyrometallurgical processes.

Large-scale non-aqueous processes to produce uranium hexafluoride, a compound unstable in the presence of water, were developed during 1943-1945; they are used routinely on a scale of tons per day to produce feed for the gaseous diffusion process used to enrich uranium. When it was found that plutonium also formed a volatile hexafluoride with characteristics very similar to those of uranium hexafluoride, it became apparent that uranium and plutonium might theoretically be recovered from irradiated fuels on an industrial scale by taking advantage of the fact that few other elements form volatile fluorides and that fluoride volatility processing might also theoretically be used to recover neptunium as NpF_6. However, volatile actinide fluorides other than uranium are unstable, and fluoride volatility processing is only practical for recovery and purification of uranium (Benedict et al., 1981).

Two basic types of molten salt systems have been used in separations. A mixed molten fluoride salt was used as a coolant, homogeneous fuel, and blanket system in the Molten Salt Reactor Experiment (MSRE) and for the reduction of the actinide fluorides to the elements (Rosenthal et al., 1972). Eutectic chloride salts are used as an ionic solvent in pyrochemical reprocessing systems that are intended for use with highly irradiated spent metallic reactor fuels

(Steunenberg et al., 1969; McPheeters and Pierce, 1992). Oxide fuels have also been reduced to metal in systems using molten lithium metal and lithium chloride, and preliminary results are available.

A technique known as plutonium electrorefining was developed (Coops et al., 1983; Christensen and Mullins, 1983) for actinide systems in the mid-1960s as a way to purify the plutonium metal alloys that were proposed as fuel for the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE) project. Electrorefining is classified as a pyrochemical process for the purposes of this discussion, since it uses a molten chloride salt as the ion transport medium between an impure metal anode and a much purer metal cathode (the collected product). The basic electrorefining technique has been adapted for purifying plutonium from essentially all elements, including americium, and for preparing pure uranium and plutonium metals for both the weapons and the breeder reactor fuels programs.

TYPES OF WASTES

Spent Reactor Fuel

By far the largest amount of radioactive waste now accumulated or currently being produced in the United States is in the spent fuel from light-water power reactors (LWRs). The principal elements in them are actinides (primarily uranium) and fission products, with the radionuclides of primary concern changing somewhat with the time frame being considered. The fission products, in particular cesium, strontium, and the rare-earth fission products, dominate as the principal sources of radioactivity and of heat for some 70 years after removal from the reactor (the activation products decay rapidly). After 300 years of decay, ^{239}Pu, ^{240}Pu, ^{238}Pu, ^{241}Am, ^{151}Sm, ^{237}Np, and ^{99}Tc are the major sources of radioactivity. After 10,000 years the major radionuclides of concern are ^{239}Pu, ^{240}Pu, ^{99}Tc, ^{237}Np, and ^{243}Am; and after 100,000 years, ^{237}Np, ^{242}Pu, ^{99}Tc, $^{234, 235, 238}$U and their decay daughters.

Defense Nuclear Waste

Separations processes are needed throughout the Department of Energy (DOE) weapons complex to decontaminate residues, equipment, and facilities. A very important goal of these operations is the removal of the hazard posed by the millions of gallons of radioactive liquids, sludges, and salts stored at the DOE defense sites where large-scale chemical-processing activities were carried out over many years. The defense nuclear wastes of greatest concern are those in liquid waste storage tanks at Hanford, although large amounts of wastes generated in the course of nuclear weapons production are stored at the Savannah River Site and the Idaho Chemical Processing Site, and small amounts from the early years are stored at West Valley Demonstration Project and Oak Ridge. The needed separations range from those achievable with basic mechanical operations to those involving sophisticated chemical processing and complex engineering equipment.

The separation and waste-form preparation systems need to be tailored to the particular situation at each DOE site. There are process-demonstration waste remediation projects under way at each site. To decide which separations processes are required for site remediation, it is necessary to know the answers to two questions that have not been adequately answered for many defense waste sites: First, what are the general composition and characteristics of the specific waste whose constituents are to be separated? Second, what degree of separation is needed for the radionuclides of concern?

The applicability of separations and transmutation (S&T) to radioactive wastes from the DOE sites depends in large measure on the masses, volumes, forms, and specific radionuclides of those wastes and on the ability of the separations technologies to isolate the waste fractions. Transmutation of the longest-lived hazardous radionuclides in the Hanford tank wastes to less dangerous shorter-lived or stable isotopes by nuclear reactions has been suggested as an alternative to burying them in a geologic repository. Nearly complete separation of each radionuclide present would be necessary prior to their destruction by transmutation. To understand whether a transmutation concept is feasible for practical implementation, it is necessary to examine whether the small concentration and low total amount of such isotopes, especially of the actinides, makes this an attractive proposition to pursue with defense wastes, or is it sufficient or desirable to remove the relatively long-lived actinide isotopes and selected fission-product isotopes (e.g., ^{99}Tc and ^{129}I) from the defense wastes before their ultimate disposition and incorporate them into stable waste forms without transmutation. At present, the defense waste problem appears to call only for the selection of an appropriate site-specific separation technique; one is being implemented at the Savannah River Site, a process is being selected at Hanford, and separations are planned for Oak Ridge as well.

Separation of selected radionuclides and other materials from wastes stored in tanks at the Hanford site is particularly important because of the potential for significantly reducing the costs of the geological disposal of the tanks' contents. Engineering and pilot plant studies for evaluating the technological and economic impacts of the different approaches to concentration and partitioning of wastes, and of preparing appropriate waste forms (e.g., vitrified products) for ultimate disposal, are not yet sufficiently detailed. For example, the evaluations must consider the impact of new waste streams from each additional operation on system-wide volume minimization, as well as the increased safety and radiological risks that accompany each additional process operation.

Chapter 5, *Defense Wastes*, gives the committee's conclusions and recommendations along with details of its view on options that should be considered for managing the wastes stored in tanks at the Hanford site.

TYPES OF SEPARATIONS

The nuclear wastes in spent LWR fuel differ markedly from defense nuclear wastes, such as those in the Hanford waste tanks. In the Hanford tank wastes the radionuclides of major interest are limited to ^{137}Cs, ^{90}Sr, ^{99}Tc, and ^{239}Pu. Also present in spent LWR fuel are americium, curium, neptunium, and many additional fission products. The processing of spent reactor fuel is among the more demanding technologies in the chemical field. The complexity results from the requirements for recovery and separation of radionuclides at high purity levels, the effects of radiolysis, and the relatively low concentrations of the species to be separated. The demands on the system under such conditions are often extreme, as the separations work must be done in high-radiation fields. The operations must also take into consideration the fact that the solutions containing concentrated actinides could conceivably produce a critical mass in the processing equipment. The amounts of materials present must be limited, and some processes need to be conducted in criticality-safe equipment. Despite the apparent degree of difficulty, nuclear fuel reprocessing technologies have been developed and deployed for both civilian and defense purposes in a number of countries. In particular, large reprocessing plants in France and the United Kingdom are now operating on a commercial (fee-for-service) basis, and smaller plants are operating in other countries. The status of nuclear fuel reprocessing is summarized in Appendix D.

To facilitate the separation of the actinides, advantage is taken of their several oxidation states. To achieve high separation factors and recoveries, the processes must use several stages in series. When the separation factors obtained in each stage are very low, it may be necessary to recover the end-products and recycle them through the system repetitively to get the necessary overall separation factor required. In this way, even processes in which the individual stages have factors of 1.1 or lower can theoretically be used to accomplish very effective separations, although the resulting cascade may be too large to be practical.

From the point of view of the principles involved, it makes little difference whether the process is conducted with aqueous and organic liquid solutions or in molten salt and metals. Pyrochemical processes might require less capital expense than aqueous ones because the volume of shielded space can be smaller, but staging could become more expensive because of costly materials used in the equipment and its reliance on batch processing. The original fuels may be liquids, slurries, or solids, and the solids may be metals or ceramics. Each requires a unique interface (head-end) step, and there will be an optimum initial processing step for each fuel.

In principle, the methods used to separate actinides and fission products in defense wastes and in spent LWR fuel are similar. The major problem with the Hanford tank wastes, however, is not with separations of the radionuclides, but rather with the bulk separation of the very large amounts of nonradioactive inorganic compounds, most notably sodium nitrate and nitrite, with lesser amounts of caustic soda, sodium carbonate, and iron hydroxide, and of organic compounds. Adaptions (sometimes involving significant development) of conventional processing and chemical engineering techniques may be used for concentrating and isolating many of these bulk materials. For example, selective dissolution and dilution can dissolve bulk components such as sodium nitrate and sodium nitrite from less soluble residues. More sophisticated techniques are required for isolating the radionuclides. Dissolution followed by solvent extraction may be used to isolate TRUs; ion exchange may be used for removing strontium, cesium, and technetium; flocculation may remove suspended trace-level colloidal plutonium species from aqueous streams. All of these processes appear to be feasible but have not been demonstrated on actual wastes from the Hanford tanks. Whether programs to develop new and better techniques are needed or not depends on pragmatic evaluation of the alternatives on a sound technical basis balancing advanced technology against economics.

Aqueous Processes

Aqueous processes have been in use for LWR spent-fuel reprocessing in several countries. They are the principal type of separations proposed for concentrating radioactive species in defense wastes (Chapter 5) and have been suggested as an option for recycling TRUs in several proposed transmutation systems.

Aqueous processes involve chemical separation of dissolved radioactive isotopes. The initial fuel dissolution process may involve very corrosive materials and require expensive equipment. After dissolution of the fuel matrix in nitric acid, the PUREX process separates $U^{(6+)}$ and $Pu^{(4+)}$ from fission product species in nitric acid solution by solvent extraction with tributyl phosphate (TBP). Americium, curium, and, under most conditions, neptunium, remain in the waste stream. Reduction of plutonium to Pu^{3+} diminishes its extractability in the TBP phase and makes it readily separable from uranium. The transuranic extraction (TRUEX) process (Horwitz et al., 1990) is also a solvent extraction system; the powerful extractant that it uses offers greater extractability of +3 actinides than does PUREX. The TRUEX process requires the PUREX process as its front end. Research and development studies suggest that it has promise for extracting americium and heavier actinides as well as residual amounts of uranium and plutonium. Ion exchange using both organic and inorganic exchangers finds wide application in radiochemical separations. However, when organic exchangers are used, special attention must be paid to radiation effects on the organic materials, because none of them is totally resistant to radiation, and the degradation products affect the operation of the extraction system.

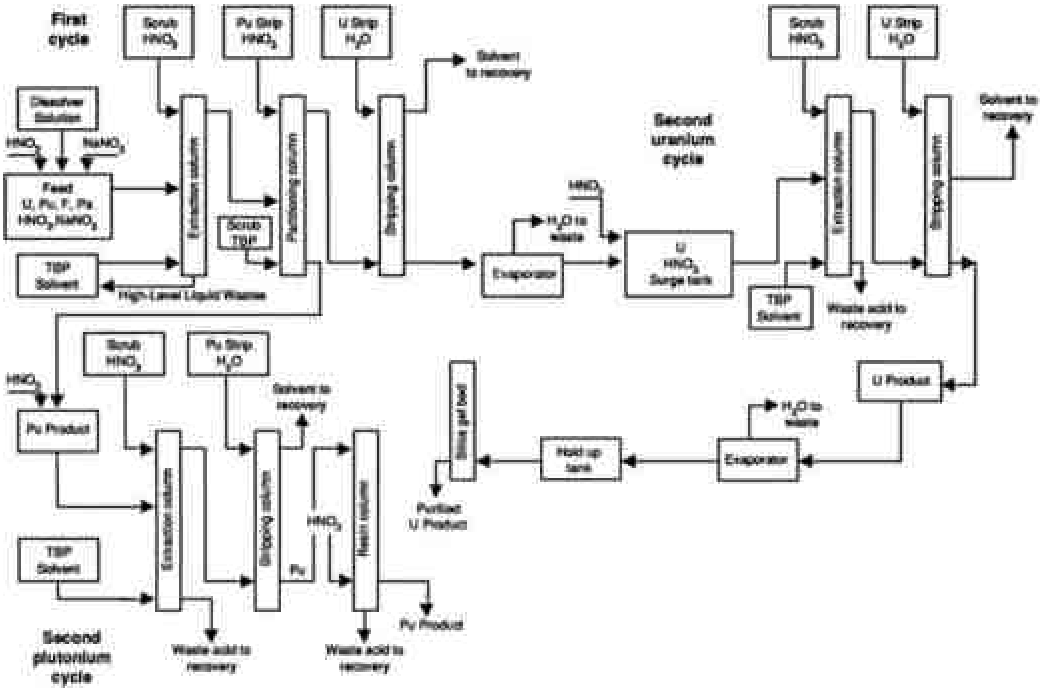

FIGURE 3-1 Flowsheet of the PUREX process. SOURCE: Keller, 1971.

PUREX Process

From the hundreds of promising separation methods studied in the laboratory, PUREX has emerged over the years as the predominant method for commercial nuclear fuel reprocessing in a number of countries, notably France, the United Kingdom, Russia, and Japan. This process, shown schematically in Figure 3-1, typically employs an approximately 30% solution of TBP in an organic diluent (normally a saturated hydrocarbon like kerosene) to extract uranium and plutonium from nitric acid solutions. The chemical basis for the PUREX process is that TBP selectively extracts uranium and plutonium when they are oxidized and in a complexant solution of high ionic strength, such as that provided by moderately concentrated nitric acid. Neptunium may also be extracted when conditions are adjusted properly. Plutonium is then selectively removed from the organic stream by contacting it with moderately concentrated nitric acid containing a reducing chemical in an operation called "stripping." The organic phase is then contacted by a stream of dilute nitric acid that returns the uranium to the aqueous phase. Usually one or two additional cycles of extraction/stripping with decontamination factors (DF)[1] between 10^3 and 10^4 are used for both the uranium and plutonium streams, resulting in overall Dfs

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[1] The decontamination factor (DF) is the ratio of the impurity concentration relative to the desired product before processing to that concentration after processing; in the case at hand, the ratio of fission product concentration in uranium or plutonium before processing to the same concentration after processing.

from fission products in excess of 10^7 for each element. Some plants employ an ion exchange step in place of the last plutonium extraction cycle, with similar results. Other separation steps would have to be added to isolate any of the individual fission product elements in the waste stream.

TRUEX Process

The TRUEX process is a solvent extraction process that was developed at Argonne National Laboratory to extract TRU elements selectively and nearly completely from acid solutions. TRUEX exists in two versions, depending on the extractant type used. Both extractants are organophosphorus compounds dissolved in hydrocarbon solvents (alkanes). One compound class, derivatives of carbamoylmethyl-phosphineoxide, is called CMPO; the other is a phosphonate called CMP. A third liquid phase[2] that is rich in the TRU elements may form when the TRU elements enter the alkane organic phase. To prevent this, TBP may be added to the organic phase. The addition of TBP prevents the formation of a third phase and enhances the extraction of the TRU elements. Third-phase formation must be avoided because its presence can cause nearly insuperable problems in the operation of continuous extraction equipment. It may sometimes be avoided by raising the temperature or by using aromatic or chlorinated hydrocarbon diluents instead of saturated hydrocarbons. Isopropyl benzene has been employed as the solvent for this reason. However, solvents other than alkanes are often considered to have unacceptable flammability and health and safety problems.

In many common solvent extraction processes, dilute nitric acid is used to back-extract or strip tetravalent actinide ions from the organic extractant into the aqueous phase. This is not practical with the TRUEX-CMPO system (the structure of the TRUEX-CMPO system is given in Appendix D) for the more strongly extracted elements because of strong chemical bonding. Several organic compounds are being studied for stripping actinides from the TRUEX solvent. Complexing agents that add more to the waste volumes to be treated are often used, and studies on how to avoid them are needed.

In addition to extracting the TRU elements, the TRUEX-CMPO solvent also extracts nitric acid from acidic feed solution, as well as other protonated species such as heptavalent technetium as in pertechnic acid. Thus, there is a possibility that the system might be adapted to remove technetium from acidic wastes. It is not yet clear whether the CMPO extractant would be near-optimal for trivalent element separations.

The TRUEX process requires further research and development and production-scale testing before it can be considered to be fully available for use.

Other Aqueous Processes

In addition to PUREX and TRUEX, there are a number of other TRU and fission-product separations processes that have, or hold promise for, application to spent reactor fuel or defense wastes. The DIAMEX process (Musikas and Hubert, 1983; Musikas and Schulz, 1992) is a solvent extraction process for TRU elements that has undergone considerable development work in France. In this process, amides, for example, dimethyldibutyltetradecylmethylamide (DMDBTDMA)[3] are dissolved in alkanes to extract TRU elements selectively, and with good separation factors, from nitric acid solutions. Advantages claimed for this process are very low solubility in nitric acid, good extraction of metal ions without third-phase formation, and good thermal and radiolytic stability. An amide with improved properties is being sought for some of the more difficult separations, such as that required for $Am^{(III)}$. A particular advantage of this class of solvents is that they can be incinerated completely, while, under ordinary circumstances, the ash inherent in the combustion of the phosphorous-based extractants is a significant waste product.

Process scientists in Russia and the former Czechoslovakia are investigating the use of cobalt dicarbollide as an extractant for cesium and strontium (Mason et al., 1990). They have developed large-scale production methods for the reagent and have recovered megacuries of cesium and strontium from actual PUREX-processed acid HLW. The extractant possesses good radiation resistance, has good selectivity for cesium and strontium, and is stable over a wide range of acidities. A potential drawback to the dicarbollide is that it must be dissolved in a polar solvent, which in turn has significant solubility in water. For applications to alkaline wastes as at Hanford tank wastes, engineering studies involving non-halogenated aromatic solvents will be required.

Ion exchange processes have had considerable success in aqueous processing for removal of cesium and strontium from wastes. They are the basis of industrial processes for the separation of individual rare earth elements and other trivalent elements like the heavy actinides. Organic ex

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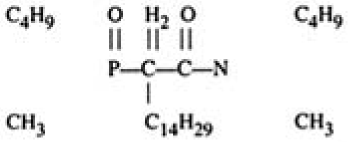
[2] Third-phase formation can be a problem in the PUREX process also, but it is controlled with less difficulty by dilution.

[3] Structure of dimethyldibutyltetradecylmethylamide (DMDBTDMA) (Musikas and Hubert, 1987).



$$\begin{array}{ccccc} C_4H_9 & O & H_2 & O & C_4H_9 \\ & \| & \| & \| & \\ & P- & C- & C-N & \\ & & | & & \\ CH_3 & & C_{14}H_{29} & & CH_3 \end{array}$$

changers show promise for selective cesium removal from alkaline waste, and inorganic zeolites such as the titanates and silicotitanates may find application in selective removal of both cesium and strontium from alkaline wastes. The reagents used for column regeneration and for elution of the desired components of the feed are considerable waste producers in many processes.

Further discussion of these and other emerging separations technologies applicable to radioactive wastes is presented in Appendix D [4] .

Non-Aqueous Processes

Pyroprocessing is a major separations technology that has proved effective in recycling actinides in defense-related (primarily plutonium-bearing) materials and metallic fuel at EBR-II. Plant designs based on preliminary research and development results indicate that pyroprocessing has the potential to reduce the size of plants and equipment needed (Steunenberg et al., 1969; McPheeters and Pierce, 1992), and thus the cost, as compared to a plant based on aqueous reprocessing. Other attributes of pyroprocessing plants such as their inherent high-temperature batch operations, when coupled with the preliminary nature of fundamental design assumptions, suggests that the indications of lower cost are very uncertain and not sufficiently reliable to form a basis for current decisions.

Another important advantage of pyroprocessing is its ability to handle fuels from the ATW and the ALMR that have been out of the reactor for much less than a year. The inventory build up is lower and the operations are on a smaller mass flow scale relative to PUREX. The nonaqueous nature of the process reduces the radiolysis problems. The high decay heat may even be an advantage since pyrochemical processing typically is performed at process temperatures of 500 to 800°C. If specifically designed for such a purpose, pyrochemical processing of spent reactor fuel can provide high separation factors between members of chemically similar families of elements. However, it requires structural materials that can withstand the high temperatures and the corrosive molten salts. A variety of processes that promise improved separations have been proposed on the basis of laboratory research. Only the major ones are mentioned here; more detail is provided in Appendix D.

Electrochemical Separations—IFR

The Integral Fast Reactor (IFR) fuel separation process, which had been under development at Argonne National Laboratory, is based on the selective electrorefining of uranium, plutonium, and heavier actinides from a molten cadmium solvent (the anode) into which they have been dissolved by anodic dissolution of spent IFR fuel. The basis of electrorefining is that, under a given set of conditions, each metallic chemical element has a well-defined, nearly unique electrical potential at which it dissolves in a medium such as molten chloride salt, allowing elements or closely related element groups to be separated. Conversely, there is also a potential at which an ionic form of a metallic element is removed, or plated, from an ionic solution. By adjusting electrical potentials appropriately, it is possible to transfer metallic elements such as uranium and transuranic actinides selectively from a metallic phase to an ionic phase and back again, and thus to effect their separation from one another and the bulk of the fission products. The IFR fuel separation process is designed to separate the transuranic actinides as a group and not produce an essentially pure plutonium stream. However, this also results in the plutonium product containing a significant amount of rare earths that reduce the worth of the fuel during recycle. The electrorefining process somewhat resembles the salt transport process described in Appendix D, but the driving potential is provided electrically rather than chemically and can be controlled with greater precision.

In the case of the IFR fuel separation process, actinide elements are dissolved electrochemically from spent fuel and deposited in a molten cadmium metal cathode. The cadmium forms an intermetallic compound, with plutonium and the heavier actinides, which increases their chemical free energy by about 17 kcal/mole. This increase is large enough to change the dissolution potentials so that uranium present in the cell (either in the cadmium phase or in equilibrium with the ionic salt solvent) is transported preferentially from the cadmium and deposited on a solid cathode. On depletion of the uranium, the operating cell voltage rises slightly; plutonium (with other actinide metals and some rare earth fission products) is then transported across the ionic salt phase to the solid cathode. In the IFR process, the cathodes are changed at the onset of a cell voltage rise, effectively accomplishing separation of uranium from plutonium electromechanically. The more chemically active elements (e.g., cesium and strontium) remain as ions in the salt phase. The more noble fission product metals (e.g., palladium, rhodium, technetium, and ruthenium) are not oxidized at the anode and remain as elemental impurities in the cadmium solvent anode. Eventually, both the cadmium and transport salt phases are processed chemically, the salt by stripping with a lithium/cadmium alloy and the cadmium by distillation, to regenerate the alloy and salt phases. The fission products remaining in the cell salt are discarded as waste.

A more complete description of these proposed processes

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[4] Alternate technologies and processes which are in the initial stages (e.g., TRPO based) have not been examined in this report.

appears in Appendix D (Appendix D-19 through D-24 and reference in the Appendix D to Y.A. Chang et al., 1992).

Molten Salt Processes—ATW

All but one of the several potential separations processes proposed by the accelerator transmutation of waste (ATW) project would use a molten mixed-fluoride salt as a carrier for the primary target loop. The molten salt would be circulated through a side stream external to the target region, and separations would be performed on the molten salt to produce targets for reintroduction into the nuclear reaction zone. Separations processes for use in the ATW concept must be very tolerant of the intense radiation fields produced by the fission products present in the target fluid because of the proposed short cooling times and consequent high radiation fields. It is doubtful that organic extractants could be used for these separations unless their process separation times could be reduced far below those achieved to date (It has yet to be demonstrated that molten salt processes can achieve all the proposed separations to the degree desired.)

The ATW-separations chemistry proposal is based on the Molten Salt Reactor Experiment (MSRE) carried out in the 1960s at Oak Ridge National Laboratory and development work for the proposed follow-on molten salt breeder reactor (MSBR). The MSBR used as its primary circuit fluid a molten salt of ^{7}Li-Bi-Th fluorides, with a melting point above 500°C, which served both as the solvent for the fissile material and as the cooling medium. It was found that the other actinides and fission products could be held in the melt in sufficient concentrations for a successful reactor system. For reprocessing it was found that some of the actinides and rare earths could be selectively removed from the melt by extraction with controlled concentrations of lithium metal in liquid bismuth followed by extraction with molten lithium chloride. The volatile fluorides were removed from the primary circuit molten salt with hydrogen fluoride and fluorine resulting in the removal and fractionation of iodine and uranium. Technetium acts much like a noble metal and exists in a lower oxidation state that is highly insoluble in the molten salt. In the MSRE it was found that ^{233}Pa could be extracted from the melt with ^{7}Li and bismuth in relatively pure form prior to decay to ^{233}U. However, research and development studies on some of these processes for other applications showed that serious problems remained to be addressed before such a system would be viable (Rosenthal et al., 1972) (Molten salt processes suffer from extremely severe corrosion on the materials used particularly in the "vicinity" of atmospheres containing water [vapor].)

Fluoride Volatility Processes

Isolation of uranium from bulk impurities or fission products by volatilization has been demonstrated to be a practical approach that could be scaled to industrial levels. The separation possibilities range from the recovery of uranium from ore concentrates (which is current practice) to the recovery of uranium from a molten salt (by using in-situ fluorination with elemental fluorine or perhaps the fluorinating agent O_2F_2) to produce volatile uranium hexafluoride. Since fluorides of other transuranium elements are not volatile and/or are unstable, some alternative processing technique would be needed to recover these transplutonium elements. A number of other elements of interest, such as molybdenum, technetium, and iodine, also can be volatilized from spent fuel, introducing some complications into this isolation method. The individual elements may be separated by fractional distillation.

SEPARATIONS FOR TRANSMUTATION OF LWR FUEL

This section considers the separations required for the major transmutation concepts that are discussed in detail in Chapter 4. Chemical processes are an integral part of any transmutation scheme to separate the radioactive components of the wastes into high purity fractions that can then be made into transmutation targets. Such targets would be irradiated in a neutron flux having sufficient intensity and energy such that the radionuclides in the targets would be either transmuted or fissioned into stable elements or isotopes with substantially shorter half-lives at an acceptable rate. It is desirable, in some cases, that neutron irradiations take place at specific neutron energy and flux levels during relatively narrow time intervals in order to fission the isotope before it decays to a less easily destroyed nuclide. This is true in particular for those transmutation concepts that have high neutron fluxes, such as accelerators. The narrow time window imposes severe requirements on the separations process. Figure 3-2 shows the neutron capture paths of importance in the build-up of actinides, both for neutron fluxes commonly attained in reactors and for the very high neutron fluxes available from accelerators.

The current amounts of actinides and fission products acceptable in the final LLW disposal matrix have been set rather arbitrarily by the proposers of various transmutation systems. For example, it is assumed that U.S. Nuclear Regulatory Commission (NRC) "Class C" waste requirements will be acceptable for the primary waste of the ATW process. However, final LLW criteria for this purpose have not been established by the NRC and there is no generally accepted separations goal. Therefore, there can be no as surance that any of the separations goals assumed for the proposed ATW system are valid. The same comment holds for all separations processes associated with proposed transmutation systems.

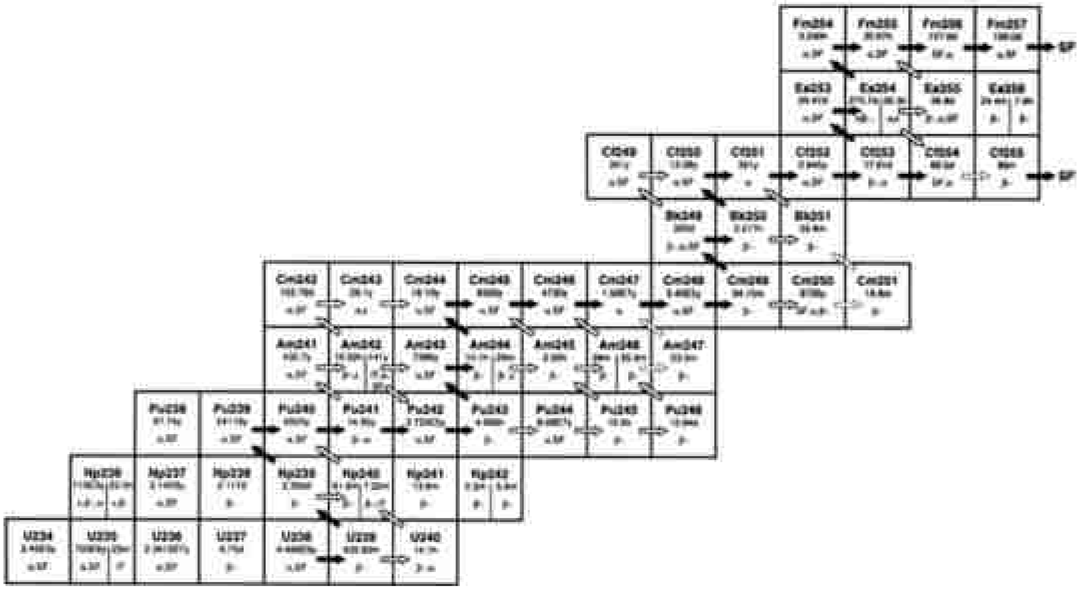

FIGURE 3-2 Neutron capture paths of importance for actinides.

The approaches being considered for S&T include transmuting and fissioning in LWRs, ALMRs, and accelerator-driven subcritical reactors (ATW) that produce about 5% of the needed neutrons by direct spallation reactions. Both aqueous and nonaqueous separations processes and their combinations have been considered for use in S&T schemes. They are described below.

Separations for Burning in LWRs

Separation of plutonium and uranium from LWR spent nuclear fuel for future use in such reactors requires the partial separation of these elements from undesirable TRUs, fission products, and zirconium-alloy cladding hulls. With minor exceptions, all existing LWR fuel reprocessing plants have employed the PUREX process with the objective of producing pure plutonium and uranium. There are no major reprocessing differences between spent fuels made with enriched uranium and those using recycled plutonium as the fissile isotope, the "mixed-oxide" (MOX) fuels. As already mentioned, the standard PUREX process does not separate americium and curium from the fission product wastes and separates neptunium only under special conditions. The modern PUREX process recovers a large enough fraction of uranium and plutonium to justify a commercially viable business and meet applicable environmental regulations (about 99.7%), which may be inadequate to meet S&T goals. To achieve the actinide recovery and separation goals that are part of many S&T concepts, and in particular to prepare targets for transmutation systems, it is necessary to augment the PUREX process with additional, much more complete separations (e.g., TRUEX).

Separations for Burning in IFRs

The high-energy neutron spectrum of the IFR (˜400 keV) makes possible its operation as a breeder to produce more fissionable material than it consumes or as a burner to fission net amounts of fissionable actinides. IFR burning has been proposed as a way to reduce present actinide inventories in spent LWR fuel. A separation methodology, either aqueous or pyroprocessing, could be used to extract plutonium and other individual actinides from spent LWR fuel for consumption in IFRs. However, a great deal remains to be done in defining important parameters such as required actinide recoveries, processes for achieving them, and costs. Periodic recycling of the IFR fuel to remove fission products and to restore the proper composition of TRU elements is required. The pyroprocessing approach that has been under investigation for this is promising; engineering-scale demonstrations of the head-end of the process have been started at Argonne National Laboratory-West in Idaho. This process, which employs direct electrochemical separation to purify the metallic reactor fuel from metal feed to metal product, could require fewer separations steps than aqueous processing when used on metallic fuels because the latter require that the metallic spent fuel be converted to an oxide or salt and then back again to metallic fuel in the course of recycling. An additional potential advantage is that it uses much smaller processing equipment because of the higher concentration of elements to be separated, although this is mitigated by the use of high temperatures, inert atmospheres, and batch processes. Further discussion of the status and further development needs of pyrochemical processes are given in Appendix D pages 14-34.

Separations for Burning in Accelerator-Driven Reactors

Both aqueous and nonaqueous separations have been proposed for accelerator-driven reactor fuels. Such reactors could transmute fission products such as ^{99}Tc and ^{129}I more rapidly than conventional reactors because they are projected to have higher thermal neutron fluxes (see Figure 3-3).

Aqueous Methods

One of the proposed ATW concepts features a target that is a slurry of TRU oxides in deuterium oxide (heavy water, D_2O) that is processed in a continuously removed side stream of the circulating target stream. This use of a fluid target and blanket material that can be processed continuously in a side stream is claimed as an advantage of the ATW system. However, there appear to be major hurdles to be surmounted in applying aqueous processes to the ATW system:

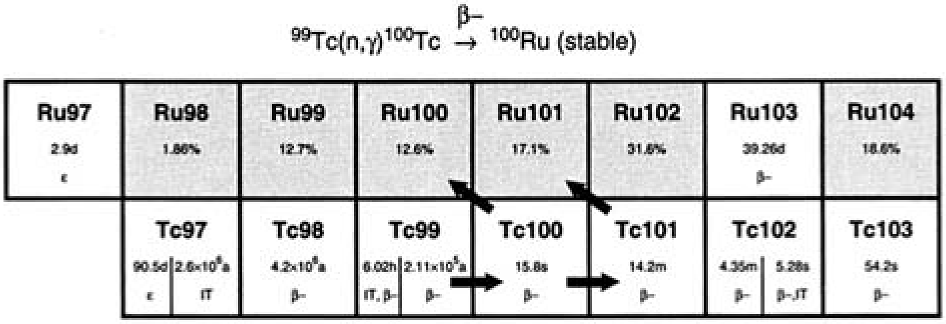

FIGURE 3-3 Transmutation of ^{99}Tc.

1. Two separate target loops would be needed, one to handle the uranium, plutonium, and higher actinide elements, and the other for the technetium and iodine.
2. Criticality concerns mandate very low inventories in the circulating slurries and in the separation plant.
3. The ATW proposal involves first evaporating the deuterium oxide fuel slurry to dryness and then dissolving it in nitric acid to allow aqueous processing with organic extraction processes.
4. The proposed use of continuous rather than batch processing may be overly optimistic due to the intense radioactivity: some interim storage is likely to be necessary to reduce the heat load resulting from the decay of short-lived radionuclides, since this heat could greatly complicate the aqueous separation processes. Radioactive decay power levels would have to be below about 10 watts per liter to prevent boiling of the aqueous solution; for instance, the heat load from any ^{242}Cm produced (120 watts per gram) would require that no more than about 0.1 grams per liter be in the processing stream.

These considerations may make the objectives of actinide fissioning and fission product destruction extremely difficult to obtain.

Brookhaven National Laboratory proposes a conventional PUREX aqueous separation system for its oxide-based accelerator system. It involves aqueous electrochemical oxidation of the carbon-coated fuel of their pebble bed reactor followed by a PUREX process. This cycle would benefit from the advanced processes that are being developed in connection with other proposed systems, as long as their fuel type can be efficiently processed and the active material dissolved in an aqueous solution. No demonstration of this process system has yet been performed.

Nonaqueous Methods

A variety of nonaqueous separations, including fluoride volatility and molten salt processes, are also under study for the ATW system. All pose challenging problems. Previous research and development on fluoride-volatility and molten-salt processes revealed several serious difficulties in developing metal alloys that would resist corrosive attack by the tellurium fission product. New techniques are needed for operating separation cascades in molten-salt and molten-metal systems to achieve some of the very high separations needed.

The separations research and development necessary for these accelerator-based transmutation schemes would be more extensive than that required by the IFRs because a wider variety of separations options must be considered. A circulating molten salt that is processed by pyrochemical techniques may be the best solution for operating in the very high neutron flux and with the short-cooled materials in the ATW system, and it is this approach that has been receiving major emphasis at the Los Alamos National Laboratory. No demonstration of this technology has yet been performed. Extensive laboratory study followed by demonstration of the technology would be required, as there has been little experimental research on the proposed ATW schemes.

Combination Methods

Combinations of aqueous and nonaqueous separation steps are also being considered by the research groups developing the ATW systems. There are special considerations at the interfaces between aqueous and pyrochemical steps involving the need to convert dissolved aqueous species to and from metals or salts, and these add to the costs of operations and waste generation.

CONCLUSIONS

For any of the S&T systems proposed, a variable but substantial number of S&T cycles would be required before the amounts of actinides and selected long-lived fission products were reduced to the target levels suggested by those developing S&T concepts. Substantial improvements and innovations in separations technology suitable for very intense radiation fields are required to obtain the high degrees of separation and low process losses per cycle needed. Special attention would have to be given to losses in side streams and to waste streams from all the parts of the fuel cycle.

More detailed findings and conclusions related to the separations aspects of the various approaches proposed are outlined in the paragraphs below. Conclusions and recommendations for defense wastes may be found in Chapter 5.

- Improved separations technologies have the potential to reduce the amounts of high-level and TRU radioactive wastes by removing nonradioactive and low-activity bulk materials (primarily uranium) from spent LWR fuel waste. The usual PUREX process has overall losses of plutonium and uranium of a few tenths of a percent, yielding waste streams that are categorized as TRU waste. However, by using a modified PUREX process for enhanced plutonium, neptunium, and uranium separations and treating the resultant waste streams with advanced separations processes such as TRUEX, it might be possible to produce a waste with a 100- to 1,000-fold reduction in those actinides. These reductions are goals that can be reached with known (but not fully developed) technologies—at a price. In addition, selected long-lived (e.g., ^{99}Tc) or highly gamma radioactive (e.g., ^{137}Cs) fission products could be removed by means of ion exchange or solvent extraction processes that have been demonstrated at the laboratory level. Such an approach would produce a very low volume of high-level and TRU wastes, and a low-level waste suitable for surface or near-surface disposal with the attendant requirement for long-term (˜300 years) storage facilities for cesium and strontium.
- Aqueous separations technologies based on new solvent systems and new ion exchange materials are in various stages of development. None has been demonstrated at the full engineering pilot-plant level. For application to spent LWR fuel processing, an advanced technology (e.g., one that can separate elements in oxidation state III) could be used following an improved PUREX process to achieve very high decontamination factors for transuranium elements.
- The Department of Energy has proposed pyroprocessing of spent LWR fuel to achieve the separations needed for transmutation in an IFR system. Pyroprocessing involving electrorefining has been demonstrated on the several-kilogram scale with metallic IFR fuel. Kilogram-scale recovery has been performed with spent LWR fuel. Although research and development is not sufficiently advanced in this area to support firm conclusions, pyroprocessing of the type being developed for the IFR appears to be a satisfactory concept for processing LWR fuel if metallic fuels are to be used subsequently.
- The ATW program goals for process losses per cycle are 0.02% or less for plutonium and neptunium and 0.0003% or less for americium and curium, for both the reprocessing of spent LWR fuel and the on-line, coupled reprocessing systems for the ATW. It is assumed that those goals are sufficient to permit waste from the ATW to be treated as LLW not requiring disposal in a geologic repository. However, a major process development program would be required to meet them. The separations processes for the ATW are substantially more challenging than those under consideration for the other transmutation concepts. The difficulties arise from two factors. The first is the very

short cooling times that are proposed before separations are performed on the irradiated targets. This time factor makes it extremely difficult to use organic reagents and aqueous systems for the major separation steps. Excessive radiolytic degradation of the organic separations agents and water radiolysis would be unavoidable. Pyroprocesses based on molten-salt and molten-metal systems are more radiation resistant and would not be subject to the same problems. The second factor is the large number of separate streams that must be processed because actinides, technetium, and iodine must all be processed separately, and recovered essentially completely, for recycling. Because of the first factor, this is likely to be accomplished with less-familiar nonaqueous processes. The ATW separations concepts that have been proposed are far short of being demonstrated processes; in fact, they are at such a preliminary stage of study (primarily on paper) that any judgment on their ultimate viability is necessarily premature at this time.

- Waste materials generated during processing and maintenance operations for the ATW would have to be treated for recovery to degrees never before achieved in large-scale systems in order to reach the low overall system losses required by the stated ATW goals.

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Chapter Four

Transmutation Systems

The previous chapter focuses on the chemical technologies for separating radionuclides in spent fuel and defense high-level radioactive waste (HLW). This chapter considers proposals for attaining the neutron fluxes that could transmute such radionuclides, focusing on the prospects for benefiting the disposition of spent fuel from civilian reactors.[1] For achieving transmutation, any practical system must integrate transmutation with separations of radionuclides to be transmuted by reprocessing of spent light-water reactor (LWR) fuel assemblies. In addition, all the proposed transmutation processes have a complex fuel cycle and would involve not one new facility but an interdependent system of many. Such systems require many components: spent-fuel reprocessing plants, waste transmutation facilities with systems to remove heat and (optionally) convert it into electricity, fuel fabrication plants, processing plants for the wastes from the separations and transmutation processes, sites for eventual disposal of the low-level radioactive waste (LLW) and any remaining HLW, as well as transportation systems for moving fuel and radioactive material between sites.

This chapter assesses the basis for transmutation by taking a systems point of view on the following proposals made by various organizations to the U.S. Department of Energy (DOE).

- Advanced liquid-metal reactor (ALMR) as part of an integral fast reactor (IFR) system, proposed by General Electric (GE) and the Argonne National Laboratory (ANL).
- Particle bed reactor (PBR), proposed by the Brookhaven National Laboratory (BNL).
- Accelerator transmutation of waste (ATW) system, proposed by the Los Alamos National Laboratory (LANL).
- Phoenix accelerator-driven fast reactor concept, proposed by the Brookhaven National Laboratory.
- Clean Use of Reactor Energy (CURE), a study[2] by Westinghouse Hanford Co. and Battelle Pacific Northwest Laboratories, involving modified liquid-metal fast reactors (LMFR) as waste transmuters in a system with LWRs.

In addition to these systems, the LWR is evaluated as a transmuter of transuranics (TRUs) and selected fission products from reprocessed LWR spent fuel. An LWR transmuter is a feasible approach in its own right, assuming the development of a fuel cycle to support such transmutation. In the analysis presented in this chapter, an LWR transmuter system serves as a reference for evaluating the advantages, disadvantages, and development requirements of the other proposals.

The chapter presents some judgments on the current level of technological maturity of the proposed systems and the prospects for their further development, using information from foreign programs for perspective. The fuel cycle required to support transmutation is considered, including separations, waste treatment, technical issues, and costs. The impacts on the repository are covered. Proposal-specific aspects of crosscutting issues from fuel-cycle economics, impacts on the repository, health and safety, licensing, and the institutional context are reviewed, but a comprehensive discussion of such issues, and of nuclear proliferation, is deferred to Chapter 6. A detailed evaluation of the concepts is provided in the appendices to this report.

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[1] Due to the low concentration of transuranics in defense HLW, it is not considered in this chapter.

[2] The CURE study did not propose the development of a specific reactor system. Rather, the study assessed the issues involved in transmutation and proposed a follow-on study to define a specific system and its technology base. The CURE study results and the issues it raises are covered in the evaluation of the other four proposals plus the LWR transmuter; a "CURE system" as such does not appear as a distinct transmutation option in this report.

The sections listed below summarize the evaluation of the three primary transmutation concepts, namely, the ALMR/IFR, ATW, and LWR systems, plus an initial assessment of the PBR and Phoenix systems, on which less information is available. The evaluation that follows in the sections to this chapter covers the following topics:

- transmutation processes and concepts;
- reduction of transuranic inventories;
- reduction of key fission product inventories;
- safety issues for the reactor, fuel materials, and supporting fuel cycle;
- development time, cost, feasibility, and risk through system demonstration;
- time scale and costs for complete system deployment; and
- comparative thermal and electrical efficiencies.

TRANSMUTATION PROCESSES AND CONCEPTS

The subsection below, *Transmutation Processes*, reviews the principal nuclear processes for transmuting the key fission products and transuranics (TRUs). The subsections *Transmutation Reactor Approaches, Overview of Critical Reactor Concepts*, and *Overview of Accelerator-Driven Reactor Concepts* introduce the transmutation proposals and the LWR as a waste transmuter and reference system for the proposals. The subsection *Separations with Transmutation of Plutonium (Only)* introduces a simplified transmutation concept in which the only TRU burned is plutonium, possibly with key long-lived fission products.

Transmutation Processes

Transmutation of Key Fission Products

The main transmutation processes for fission products are neutron capture (producing a higher mass nuclide) and beta decay. For many fission products the neutron capture cross sections in a thermal (or epithermal) spectrum can give substantial transmutation rates. The corresponding capture cross sections in a fast neutron spectrum are typically orders of magnitude smaller.

Transmutation of the long-lived fission products ^{99}Tc and ^{129}I is feasible in a thermal reactor. The ruthenium and xenon transmutation products are stable under neutron capture processes; that is,

$$\begin{array}{lllll} {}^{99}\mathrm{Tc} + \mathrm{n} \rightarrow & {}^{100}\mathrm{Tc} \rightarrow & {}^{100}\mathrm{Ru} + \mathrm{n} \rightarrow & {}^{101}\mathrm{Ru} + \mathrm{n} \rightarrow & {}^{102}\mathrm{Ru} \\ & (16\ \mathrm{s})\beta & \text{stable} & \text{stable} & \text{stable,} \end{array}$$

and

$$\begin{array}{lllll} {}^{129}\mathrm{I} + \mathrm{n} \rightarrow & {}^{130}\mathrm{I} \rightarrow & {}^{130}\mathrm{Xe} + \mathrm{n} \rightarrow & {}^{131}\mathrm{Xe} + \mathrm{n} \rightarrow & {}^{132}\mathrm{Xe} \\ & (12.4\ \mathrm{h})\beta & \text{stable} & \text{stable,} & \text{stable,} \end{array}$$

and

$$\begin{array}{lllll} {}^{127}\mathrm{I} + \mathrm{n} \rightarrow & {}^{128}\mathrm{I} \rightarrow & {}^{128}\mathrm{Xe} + \mathrm{n} \rightarrow & {}^{129}\mathrm{Xe} + \mathrm{n} \rightarrow & {}^{130}\mathrm{Xe} \\ \text{stable} & (25\ \mathrm{min})\beta & \text{stable} & \text{stable} & \text{stable.} \end{array}$$

The ^{129}I is produced with stable ^{127}I, which also transmutes to stable isotopes. For a thermal neutron spectrum typical of a uranium-fueled PWR, the spectrum-averaged capture cross sections for dilute ^{99}Tc and ^{129}I from the ORIGEN-2 code library are 13.8 barns and 3.2 barns, respectively (Wachter and Croff, 1980).[3] An ORIGEN-2 calculation for a PWR gives in-reactor transmutation rates of about 11 percent per year for ^{99}Tc and 3 percent per year for ^{129}I (Wachter and Croff, 1980). Practical transmutation rates will be low because the external inventory of ^{99}Tc and ^{129}I in reprocessing and target fabrication must be taken into account.

Transmutation of ^{90}Sr (29 yr) and ^{137}Cs (30 yr) is possible in principle but not in practice. Their transmutation would make little improvement in the calculated radiological risk for a geologic repository because of the relatively short-term nature of their radioactivity, although the reduction in repository heat loading might be marginally advantageous.

Transmutation of the long-lived radionuclide ^{135}Cs would also be possible in principle but not in practice. This radionuclide would be transmuted to stable ^{136}Ba through short-lived ^{136}Cs. Unfortunately, ^{135}Cs is produced with a larger quantity of stable ^{133}Cs, which would undergo neutron capture to ^{134}Cs and to more ^{135}Cs. To remove the stable ^{133}Cs beforehand by isotopic separation would be formidable in the presence of the intense ^{137}Cs gamma radiation field. Rather than attempting to transmute ^{135}Cs, its radiological risk in a geologic repository could be reduced by separating cesium in the reprocessing operation and incorporating it into waste form in which cesium would have a significantly reduced solubility in groundwater.

Transmutation of Transuranics

Characteristics of Thermal and Fast Neutron Processes. Transmuting the TRUs is more complex (Benedict at al., 1981). In a neutron flux, several competing processes determine the concentrations of the transmuted isotopes as a function of time. Three processes are important here: (1) neutron-induced fission, (2) neutron capture to produce a higher-mass nuclide, and (3) radioactive decay. Table 4-1 lists the half-lives of selected uranium and TRU isotopes that occur during transmutation and the ratios of their spectral-averaged capture and fission cross sections for several kinds of thermal and fast neutron spectra. A number of characteristic features may be seen by a study of the properties.

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[3] 1 Bam = 10^{-28}m^2.

First, the "capture-to-fission" ratios for thermal neutron spectra show marked differences for odd-versus even-neutron isotopes. Odd-neutron isotopes fission well in a thermal spectrum; in fact, such fission is favored over capture by factors of 2 to 10. In contrast, most even-neutron isotopes are not fissioned by neutrons below several hundred kilovolts in energy, due largely to the extra nuclear stability conferred by neutron pairing. Hence, in a thermal reactor, such nuclei are fissioned only by the fission spectrum neutrons.

Second, the thermal neutron "capture-to-fission" ratios are typically higher than those for fast spectra. This effect is exaggerated for even-neutron isotopes; indeed, ^{240}Pu and ^{241}Am have large thermal neutron capture cross sections and are parasitic absorbers in thermal reactors. However, odd-neutron isotopes also show the effect by factors of 1.5 to 3, mainly because capture cross sections are typically higher for thermal spectra. Most importantly, the high thermal capture-to-fission ratio of ^{239}Pu and its capture products result in relatively large amounts of higher-mass actinides in a thermal-neutron transmuter, as compared to a fast reactor.

Transuranic Production and Transmutation. The main source of the principal TRU, ^{239}Pu, is neutron capture in ^{238}U followed by two beta decays (i.e., in the initial spent fuel plus possible additional production during transmutation). The ^{239}Pu fissions well with neutrons of any energy. Alternatively, it produces ^{240}Pu by neutron capture. Table 4-1 lists a capture-to-fission ratio of 0.55 in a thermal spectrum compared to 0.17 and 0.26 in a fast spectrum with metallic and oxide fuel, respectively. Successive neutron captures, starting with ^{239} Pu, produce higher mass isotopes of Pu, Am, and Cm.

The minor actinides and other plutonium isotopes, in particular ^{240} Pu and ^{242}Pu, fission well in a fast spectrum. That isotope ^{237}Np could undergo neutron capture and furnish additional ^{239}Pu. That is,

$$^{237}\mathrm{Np} + \mathrm{n} \rightarrow \underset{(2.1\ \mathrm{d})\ \beta^{-}.}{^{238}\mathrm{Np} \rightarrow} {}^{238}\mathrm{Pu} + \mathrm{n} \rightarrow {}^{239}\mathrm{Pu}$$

However, in a very high neutron flux, the intermediate nucleus ^{238} Np could attain a high probability of capturing a second neutron and fissioning before the beta decay could take place. Thus, in principle, a high-flux transmutation scheme could achieve a higher fissioning rate of various isotopes than a scheme that operates at ordinary thermal flux levels.

Two even-neutron isotopes in the plutonium chain, ^{240}Pu and ^{242}Pu, are key precursors of higher-mass actinides by neutron capture to ^{241}Pu and ^{243}Pu, which can undergo beta decay to ^{241}Am and ^{243}Am, respectively (see Table 4-1). The two americium isotopes fission well in a fast spectrum, but can produce ^{242}Cm (163 d) and ^{244}Cm (18.1 yr) by neutron capture in a fast or thermal spectrum. However, the higher capture-to-fission ratio characteristic of a thermal spectrum, discussed above, results in a build-up of higher-mass actinides during thermal neutron transmutation. A chain of long-lived curium isotopes is produced by successive neutron captures—^{245}Cm (8,500 yr), ^{246}Cm (4,820 yr), ^{247}Cm (1.56×10^{7} yr), and ^{248}Cm (3.7×10^{5} yr), even some ^{250}Cm (9.7×10^{3} y). The chain branches at ^{249}Cm with the 64-minute beta decay to ^{249}Bk, which leads in steps to production of californium isotopes, in particular ^{252}Cf. Even higher-mass actinides can be produced in a thermal flux $>10^{15}$ neutrons/cm^{2}-s, such as in the high-flux isotope reactor (HFIR). Thus, a range of higher-mass isotopes is produced in either a fast or thermal spectrum, although the relative proportions are quite different. The possible effects of the higher actinides are discussed in the sections on several of the transmutation options. In target waste material, ^{252}Cf would be a potent source of neutrons by spontaneous fission. In addition to alpha emission, spontaneous fission also occurs in curium isotopes, becoming more probable with increasing curium mass, i.e., ^{242}Cm (8×10^{-6}%), ^{244} Cm (1.3×10^{-4}%), ^{246}Cm (0.027%), ^{248}Cm (0.83%), and ^{250}Cm (˜99%) (Hoffman et al., 1992). In addition, unburned ^{238}Pu can be an important source of neutrons by (α, n) processes. Hence, for thermal neutron transmutation concepts, significant neutron emission could present problems during fuel reprocessing and refabrication, quality assurance, and performance verification. To a lesser extent, the issue could arise with fuel rods for transmutation in a fast reactor.

Transmutation Reactor Approaches

Classes of Reactor Concepts

To take possible advantage of nuclear processes for transmutation, two quite different classes of reactors have been studied for neutron generation. The work to date consists primarily of a conceptual analysis of the effects of the neutrons generated in either approach in order to estimate the benefits and hazards that would result from changing the character of the waste to be disposed. The two classes of reactor concepts are:

1. *Critical nuclear reactors:* The nuclear assembly, containing the waste and possibly additional fissile material, operates with a net neutron multiplication factor of unity. This class includes thermal reactors, such as the LWR and PBR, and fast reactors, such as the ALMR.
2. *Accelerator-driven nuclear reactors:* The nuclear assembly operates with a neutron multiplication factor less than unity, i.e., *subcritical* , so that neutrons must be added from a source external to the nuclear assembly. Intense

beams of very high-energy protons would be focused on targets such as lithium, tungsten, molten lead, or even the fuel itself. This would generate large numbers of spallation neutrons that would be multiplied by the subcritical assembly to transmute waste material surrounding the target.

TABLE 4-1 One Group Capture-to-Fission Cross-Section Ratios of Transmutation Isotopes for Thermal and Fast Neutron Spectra in Various Reactor Cores

Isotope	Half-Life	U-PWR[a]	Pu-PWR[b]	LMFBR[c]	ALMR[d]	Main Source
^{235}U	7.04 × 10^{8} yr	0.22	0.28	0.27	0.25	natural uranium
^{236}U	2.34 × 10^{7} yr	—	—	—	—	neutron capture by ^{235}U
^{237}U	6.75 d	—	—	—	—	neutron capture by ^{236}U & ^{238}U
^{238}U	4.47 × 10^{9} yr	9.02	7.93	5.44	4.34	natural uranium
^{239}U	23.5 min	—	—	—	—	neutron capture by ^{238}U
^{237}Np	2.14 × 10^{6} yr	61.3	42.43	4.22	2.71	6.8 d beta decay of ^{237}U
^{238}Np	2.1 d	0.10	0.10	—	—	neutron capture by ^{237}Np
^{239}Np	2.35 d	—	—	—	—	23.5 min beta decay of ^{239}U
^{238}Pu	87.7 yr	14.06	7.51	0.60	0.44	2.1 d beta decay of ^{238}Np
^{239}Pu	2.44 × 10^{4} yr	0.55	0.56	0.26	0.17	2.35 d beta decay of ^{239}Np
^{240}Pu	6,560 yr	178.1	70.55	1.25	0.82	neutron capture by ^{239}Pu
^{241}Pu	14.4 yr	0.33	0.31	0.18	0.15	neutron capture by ^{240}Pu
^{242}Pu	3.75 × 10^{5} yr	76.43	47.78	1.42	0.93	neutron capture by ^{241}Pu
^{243}Pu	4.98 h	0.49	0.49	—	—	neutron capture by ^{242}Pu
^{241}Am	433 yr	94.2	63.96	4.95	3.56	14.4 yr beta decay of ^{241}Pu
^{242}Am	16 h	—	—	—	—	neutron capture by ^{241}Am
^{242}Am	141 yr	0.21	0.20	0.10	0.07	neutron capture by ^{241}Am
^{243}Am	7,370 yr	106	78	3.84	2.29	4.98 h beta decay of ^{243}Pu and neutron capture by ^{242}Am
^{242}Cm	163 d	10.38	11.4	1.59	0.85	16 h beta decay of ^{242}Am
^{243}Cm	28.5 yr	0.12	0.13	0.09	0.07	neutron capture by ^{242}Cm
^{244}Cm	18.1 yr	15.79	14.83	1.71	1.21	10 h beta decay of ^{244}Am
^{245}Cm	8,500 y	0.17	0.17	0.12	0.10	neutron capture by ^{244}Cm
^{246}Cm	4,820 y	5.04	4.71	0.72	0.47	neutron capture by ^{245}Cm
^{247}Cm	1.56 × 10^{7} yr	0.65	0.63	0.16	0.12	neutron capture by ^{246}Cm
^{248}Cm	3.4 × 10^{5} yr	8.36	7.88	0.68	0.44	neutron capture by ^{247}Cm
^{249}Cm	64 min	0.03	0.03	—	—	neutron capture by ^{248}Cm
^{250}Cm	9,700 yr	—	—	—	—	neutron capture by ^{249}Cm
^{249}Bk	523 d	807.0	460.3	4.63	1.92	64 min beta decay of ^{249}Cm
^{250}Cf	13.1 yr	513.6	292.8	0.37	0.20	fast beta decay of ^{250}Cf
^{251}Cf	800 yr	0.48	0.45	0.12	0.10	neutron capture by ^{250}Cf
^{252}Cf	2.65 yr	0.47	0.41	0.39	0.32	neutron capture by ^{251}Cf

[a] Normal low-enriched uranium in a pressurized water reactor.
[b] Self-generated plutonium recycle in a pressurized water reactor.
[c] Mixed oxide fuel in a liquid-metal fast reactor.
[d] Metallic fuel in an advanced liquid-metal (fast) reactor.

SOURCES: U-PWR and Pu-PWR from ORIGEN 2.1 (1991). LMRBR and ALMR from GEFR-00898 (1991).[4]

The various transmutation concepts differ widely in the many technical, programmatic, economic, and other dimensions by which they may be characterized. However, there are no universal figures of merit for the evaluation of the different transmutation approaches. The remaining sections of this chapter summarize the results of the evaluation using a variety of "measures" for comparison:

- the rate and time for various percentage reductions in the TRU inventory:
- the flexibility and rate for reducing key fission product inventories:

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[4] Other models are available for calculations and may give different values.

- the safety issues for the reactor, fuel materials, and supporting fuel cycle;
- the development time, cost, feasibility, and risk through complete demonstration;
- the estimated time scale and costs for complete deployment, including overall fuel cycle economics; and
- the comparative thermal and electrical efficiencies per net amount of waste transmuted.

The first type of measure, the reduction in TRU inventory, raises a methodological issue. Over the life cycle of a reactor, approximately 30 years, most of its initial load of TRUs is transmuted; however, additional TRUs are generated. Thus, only a partial net reduction occurs over each reactor life cycle, the untransmuted TRUs being passed to the next generation of reactors for further reduction. Some groups and authors emphasize the time to reprocess and burn the TRUs originally in the LWR spent fuel, which is much shorter than the time required for a net reduction including the TRUs created during transmutation. Others emphasize the TRUs in waste sent to a repository from reprocessing, discounting surface-stored waste at fuel-cycle facilities on the grounds that the fuel-cycle material still contains fissionable resources. However, for the major part of the transmutation period, most of the inventory is in surface facilities. This results in increased risk to nearby populations, compared to the direct disposal of spent fuel in the repository, due to the additional operations to burn the waste.

As one figure of merit for each type of transmutation system, therefore, this report evaluates the *net* decrease in *total* TRUs versus time for multiple reactor life cycles of that system, compared to an untransmuted reference case. That is, the net *TRU ratio* as a function of time is defined to be the TRUs from a once-through, uranium-fueled LWR cycle for a given electrical power level, divided by the total TRUs for the system in question (in the reactor, the associated fuel-cycle facilities and the waste) for that power level. The time scale for achieving various percentage reductions, the asymptotic reduction factors, and the neutron efficiencies for the concepts are useful for comparisons.

The calculated TRU ratios and the corresponding times for various fractional changes in net inventory range widely among the proposals. For evaluation purposes, this report considers two nuclear power scenarios, under the assumption that the transmutation systems would produce electrical power for distribution to the electrical grid. For example, for a continuing "steady nuclear power" scenario with a mix of breeding and nonbreeding transmuters, achieving a net TRU ratio of a factor of 100 requires from a few hundred years for some proposals to several thousand years for others. A TRU ratio of 10 requires from about 7 years to over 400 years for the various proposals at constant nuclear power. The time scales are significantly shorter for a "declining nuclear power" scenario, in which nuclear power is phased out as rapidly as possible consistent with transmuting the most nuclear waste. For this type of scenario, for example, the calculated time to achieve a TRU ratio of 100 ranges from about 40 years to several hundred years.

Engineering and Materials Issues

In both the critical reactor and accelerator-driven subcritical reactor approaches, there are variations in the chemical and physical properties of the fuel containing the waste to be transmuted. For this reason, all the proposals entail fuel development and verification issues. Moreover, even for the systems operating with subcritical reactors, the heat generation rates would require careful analysis of the provisions for safe removal of residual heat generated from the decay of fission products and minor actinides after the termination of the fission process. Also, the transmutation systems that employ a high fissile specific power (i.e., the thermal-reactor concepts) are likely to raise safety issues associated with the control of reactivity and power distributions in the reactor core. Detailed analysis of such issues is important for the licensability of the system designs. All the concepts require development and testing of at least some aspects in a pilot-scale system. Indeed, most of the systems will require demonstration at full-scale prototype before such systems could be implemented confidently for reliable operation at high-capacity factor.

Several proposals aim at producing and sustaining a very large fractional destruction rate—the product of neutron flux and cross section—since that determines the rate of transmutation of the waste. A large destruction rate, in turn, entails high heat generation rates from the high fission rate and decay heat of nuclei in the reactor proper and, in the accelerator-driven case, from proton deposition in the target. For these proposals the engineering designs pose difficult problems. In some proposals the materials must withstand conditions beyond engineering experience with present-day reactors. These conditions include neutron (and in the case of accelerator-based proposals, proton) fluxes and fluences, neutron spectra, temperatures, and chemical environments, singly or in combination. High-energy neutron and proton fluxes impart atomic displacement damage that generally degrades structural materials and affects basic properties (e.g., ductility, fracture toughness) and imparts dimensional instabilities (e.g., swelling and creep). Such flux levels would also increase the level of activation products in the nuclear waste.

Indeed, some of the proposed systems envision neutron fluences for certain components that are beyond the levels achieved for the corresponding components in existing systems, e.g., more than 100 displacements per atom for some structural components or roughly an order of magnitude more than experienced in the Fast Flux Test Facility (FFTF). The displacement damage can be aggravated by nuclear transmutation that converts existing alloying elements to impurities. Two such processes are (1) transmutant gas production (e.g., helium from (n, α) production) arising from the high-energy portion of the neutron spectra in accelerator-based sources, and (2) transmutation of existing alloying elements to deleterious activation products by neutron absorption in approaches with a thermal spectrum. These types of induced impurities compound the difficulties imposed on structural materials required to operate at the high temperatures due to the high flux/power and density/thermal efficiency requirements of several of the transmuter concepts. Additional deterioration can be anticipated with those concepts that employ nonaqueous chemical environments (e.g., molten salts, liquid metals, aqueous slurries) to achieve the system performance requirements. Thus, several proposals entail considerable research and development before the chances of success can be ascertained.

Program Funding

As yet, there is no focused overall program in the United States for development of S&T technologies. The objectives and time table for such an effort are difficult to work out in the present climate of uncertainty about the future of nuclear power in the United States—in particular, uncertainty concerning the prospective geologic repository at Yucca Mountain and the possible future commercialization of a fast breeder reactor sometime in the next century. This committee, therefore, is able to estimate only an approximate total expenditure level for the U.S. effort.

For FY 1992, DOE's Office of Nuclear Energy (DOE/NE) identified direct expenditures of about $19 million specifically for transmutation objectives: about $15 million for the LANL's ATW effort and about $4 million for transmutation applications of the ALMR and IFR concept. In addition, the national laboratories used discretionary funding to extend their efforts. A few million dollars was spent at LANL and BNL in FY 1992 on accelerator-based concepts for transmutation. Smaller efforts were conducted by Westinghouse Hanford and Oak Ridge National Laboratory (ORNL).

However, DOE was spending a much larger amount for research and development indirectly related to transmutation objectives. For example, the ALMR effort at GE and the IFR at ANL were aimed primarily at breeder reactor development for future power production which are both adaptable to transmutation. This support included the support for ANL-West located at the Idaho National Engineering Laboratory. In addition, the development of accelerators for tritium production and other military objectives was indirectly relevant. For FY 1992 then, the efforts directly or indirectly applicable to transmutation systems amounted to about $75 million.

The total funding allocated to separation and transmutation in FY 1993 was is $133.5 million. DOE/NE identified direct expenditures of $41.4 million specifically for transmutation objectives: $8.9 million for ALMR development at GE, $26 million for metal fuel and IFR development at ANL, and $6.5 million for LWR actinide burning. The related ANL-West supporting facilities would add $76.1 million to the FY 1993 transmutation commitment. In addition, the DOE defense budget includes $4 million for ATW-related efforts at LANL, and the waste management budget includes $12 million for efficient separations and processing.

In FY1994 the administration began a phase-down or phase-out policy of the DOE/NE programs related to separations and transmutation.

In FY1995, a total of $104.8 million has been identified for these programs including IFR, but these funds are exclusively designated for close out and termination costs.

International Activities

International activities in S&T as a method of ameliorating high-level waste management are being conducted by individual countries as well as under the coordination of international organizations (the Organization for Economic Cooperation and Development's Nuclear Energy Agency, the International Atomic Energy Agency, and the Commission of the European Communities). Most of this activity is in Japan and Western Europe, with the majority of the European activity in France. Unlike the U.S. S&T program, however, the interest in Japan, France, and the United Kingdom stems from a larger interest in commercially reprocessing fuel, both in the near-term for LWRs using mixed-oxide (MOX) fuel and in the long-term for a breeder reactor economy. As a result, these countries are working on the technologies necessary for any S&T waste management scheme, including enhanced reprocessing techniques, remote fuel fabrication, and reprocessed waste packaging.

The interest in these countries in S&T for waste management has increased recently, largely as a result of increasing public resistance to high-level waste repository siting. Japan has perhaps the largest financial commitment to this effort through its OMEGA Project, which is a research and development effort aimed at partitioning actinides from HLW and transmuting actinides in either critical reactors or accelerator-driven subcritical assemblies. French and U.K. efforts have looked at actinide burning in fast reactors, but more from a perspective of controlling reactivity swings. Coordinated European programs have considered actinide burning in fast reactor blankets, and preliminary work at the Paul Scherrer Institute in Switzerland has considered accelerator-driven spallation for actinide burning. Finally, both Japanese and European programs are examining nonaqueous separations techniques as well as enhanced aqueous reprocessing.

Overview of Critical Reactor Concepts

This subsection gives an overview of a fast reactor system and two types of thermal reactor systems that are proposed or are being considered for transmutation of nuclear waste, namely,

- transmutation of TRUs in an ALMR as part of an IFR proposed by GE and ANL;
- transmutation of TRUs and fission products in LWRs, either existing designs adapted for the purpose or more advanced designs currently in certification review by the Nuclear Regulatory Commission (NRC); and
- transmutation of minor actinides with a PBR, as proposed by BNL.

The concept of transmutation using critical reactors has been studied for several decades; indeed, the United States was a leader in the field in the 1970s. Early in the commercialization of LWRs in the United States, the U.S. Atomic Energy Commission (AEC) and the nuclear industry expected that the cost of mining and enriching uranium would increase as high-grade uranium ore supplies were used up. In the 1960s and early 1970s, the AEC projected that this would occur by the 1980s and become increasingly serious in the 1990s. The parties visualized that spent LWR fuel would be reprocessed commercially and the plutonium and unburned uranium separated and recycled, first into LWRs and later into breeder reactors when they became commercialized. Thus, the total plutonium in the fuel cycle would be limited.

Fast Critical Reactor Concepts

The ALMR/IFR program had been sponsored by DOE/NE during the previous administration under joint development by GE and ANL.[5] It emphasized the power reactor, innovative, small module (PRISM) fast reactor concept, which has been aimed primarily at the development of a breeder reactor for power production that would produce more fissile material than it consumes. It has long been recognized that a fast reactor can be modified to operate at a conversion (breeding) ratio, ß, less than 1.0 and thus could be configured to operate as a net burner of transuranics.[6] For that matter, the neutron spectrum of a fast reactor can be softened, as proposed, for example, in the CURE concept, and gain capability to transmute fission products while retaining good characteristics for transmuting transuranics.[7]

This subsection discusses the GE and ANL proposal for an IFR for burning the TRUs. An IFR would comprise an ALMR for the purpose of burning TRUs together with its own self-contained capability for processing and fabricating the metallic fuel/waste alloy. For such reprocessing, ANL proposes to use pyrometallurgical technology. ANL is also exploring pyroprocessing of LWR spent fuel to provide the TRUs to start the ALMRs; this capability might be collocated with each IFR or configured in a larger, centralized facility to support many IFRs. As an alternative, GE has considered a centralized capability for LWR spent-fuel reprocessing based on aqueous plutonium and uranium extraction and recovery by transuranic extraction (PUREX-TRUEX) technologies.

More than a dozen fast breeder reactors have been operated in the United States, the Soviet Union, the United Kingdom, France, and Japan, although none are yet on-line as breeders for reliable power production. Further development would be needed to burn minor actinides (MA), including operation of a prototype integrated system. Designs with a breeding ratio, β, of 0.22 to 1.25 appear in the ALMR/IFR project literature (Chang, 1991a, 1991b; M.L. Thompson, private communication, 1991). Recent GE studies (Thompson, 1992) involve elimination of external blankets and shortening of core height, without changes in fuel rod diameter or number of fuel rods. Preliminary ANL designs (Johnson et al., 1990) include IFRs fueled entirely with TRUs. One design involves a mixture of plutonium and MA with $\beta = 0.22$; another involves mostly MA with $\beta = 0.85$. However, with reduced fissile breeding and heavy metal inventory in the core, the TRU burner designs entail increased specific power and higher reactivity swing over a fuel cycle.

GE suggests that for a TRU burner, $\beta = 0.60$ is the minimum

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[5] In 1994, following recommendations by DOE, the ALMR/IFR was canceled by Congress. Limited funds were appropriated for phase out and for continuation of research on separations.

[6] One should note that a fast reactor with ß=1, a breeder, burns the original TRUs with which it is loaded and can properly be thought of as a waste burner.

[7] The CURE concept (Rawlins et al., 1990) assessed a modified sodium advanced fast reactor (SAFR), which has a 900 MWt (400 MWe) capacity and a homogeneous core with both radial and internal uranium blanket assemblies. The radial blanket assemblies were assumed to be replaced by hydride-modified assemblies containing the target wastes to be transmuted. The hydride would moderate the fast neutrons leaking from the core into the epithermal energy range. The spectrum is soft enough to achieve annual destruction rates for the fission products ^{99}Tc and ^{127}I of about 5 percent/yr and 10 percent/yr, respectively. Yet, the spectrum is still hard enough to burn actinides quite efficiently. The resulting breeding ratio would be about 0.8, compared to about 1.05 for the normal SAFR; thus the design is a net consumer of plutonium.

value with acceptable safety features. This limits the waste inventory reduction capability of the system. The consequences of this limitation for the inventory reduction factor, and the time required to burn various fractions of the actinides, are explored in detail later in this chapter (see Appendix F).

Thermal Critical Reactor Concepts

Several earlier studies of transmutation (Croff et al., 1977; Croff, 1980; Wachter and Croff, 1980) included LWRs as transmutation devices. Those studies focused on transmuting the MA, assuming that the recovered uranium and plutonium from LWR spent fuel would be recycled to the LWRs. Wachter and Croff also showed that an LWR could transmute ^{99}Tc and ^{129}I. However, the reprocessing of commercial reactor fuel did not go forward in the United States and is not currently considered viable for recycle to LWRs or breeder reactors, for both economic and public policy reasons. Consequently, transmutation schemes now proposed to assist the geologic disposal of LWR spent fuel (and/or accomplish other energy objectives) must deal with the transmutation of all the TRUs, the principal one being plutonium.

The section, *The Light Water Reactor* in Appendix F, discusses the LWR as a reference case for burning plutonium, the MAs, and the key fission products ^{99}Tc and ^{129}I.

The section, *The Particle Bed Reactor Concept* in Appendix F, evaluates the BNL proposal of a PBR for transmutation (Van Tuyle, 1992). The PBR is a thermal reactor concept based on a gas-cooled space reactor design. The PBR would have a compact core and operate with a power density of 5 MWt/liter, which is a factor of 10^2 greater than that of a typical LWR and even higher than the 1.13 MW/liter attained by the high flux isotope reactor (HFIR). The fuel material is embedded in small graphite particles (0.8 mm in diameter) packed in cylindrical pressure tubes. The tubes accommodate the packed particle bed within two coaxial porous plugs (frits) so that the particles could be cooled by forced circulation of helium. Heavy water and beryllium carbide moderators are being studied. The BNL proposal envisions burning the MAs, with the unburned plutonium and perhaps the unused fissile uranium to be directed for recycle to LWRs. A power reactor with such a high power density would be without precedent or experience as a commercial power reactor.

Overview of Accelerator-Driven Reactor Concepts

An alternative to using fast or thermal critical reactors is to generate a fraction of the neutrons by spallation with high-energy protons from an accelerator. LANL and BNL have made conceptual studies for achieving subcritical accelerator-based transmutation (see Appendix F). These concepts envision the production of electricity, part of it to power the accelerator and part available for sale to offset the costs of such an approach. The accelerator-based concepts are relatively new and are less well characterized and in a less mature state of technical development than the approaches using critical reactors. For these reasons, after introducing the specific proposals, the general features of accelerator-based approaches compared to those of the more familiar critical-reactor-based transmutation will be highlighted.

Appendix F treats four concepts for ATW under review by LANL for commercial LWR waste (Arthur, 1992a).[8] Spallation neutrons are produced when the proton beam strikes a lithium (or heavy metal) target inside the assembly, which moderates the neutrons to a thermal spectrum and multiplies them by a large factor, namely, 20 in the current designs (corresponding to a neutron reproduction factor $K_{eff} = 0.95$ for the subcritical assembly). The concepts differ in the type of fluidized waste/fuel (either a heavy-water slurry or a molten salt solution that circulates in a blanket), the type of processing used (aqueous or nonaqueous), the target material, and whether thorium is used in the molten-salt cases to provide production of fissile ^{233}U. Each facility would have its own on-line fuel-processing capability, various storage and waste treatment facilities, and heat conversion and electrical power generation equipment. The four concepts are as follows:

1. transmutation of TRUs and some fission products recovered by aqueous reprocessing of LWR spent fuel, using an aqueous neutron multiplying system that generates electrical power (Case ATW-1);
2. transmutation of TRUs and some fission products recovered by nonaqueous reprocessing of LWR spent fuel, using a nonaqueous neutron multiplying system that generates electrical power (Case ATW-2);
3. transmutation of TRUs and more fission products recovered by nonaqueous reprocessing of LWR spent fuel, using a nonaqueous neutron multiplying system, fueled partly with thorium and generating electric power (Case ATW-3); and
4. transmutation of TRUs and some fission products in a nonaqueous thorium-breeder system for electrical power generation (Case ATW-4).

The LANL effort is in an early conceptual design stage. The baseline design, referred to as Case ATW-1 in this report, employs known technology to the extent possible and

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[8] LANL has proposed a fifth concept for application to defense wastes, using an aqueous multiplying system similar to that of Case ATW-1. The scheme is marginal because of the concentration of TRU in the defense wastes. The concept is not further considered in this chapter.

furnishes a reference materials balance. LANL makes no claim that this design is optimized. LANL is also exploring three nonaqueous approaches with improved performance and economy. Referred to in this report as Cases ATW-2, ATW-3, and ATW-4, they attempt to optimize both the system neutron economy and overall system electric power production.

In contrast, the Phoenix concept proposed by BNL is envisioned to be part of a larger scheme for separating and transmuting waste from LWRs (Van Tuyle, 1992). As discussed in the section, *The Accelerator-Based Phoenix Concept* (see Appendix F), the Phoenix proposal would transmute the MAs and the radio-iodine from the reprocessed LWR spent fuel. The separated plutonium and uranium would be recycled to LWRs. Like the LANL ATW concept, Phoenix generates neutrons by spallation with a high-energy proton beam from an accelerator and multiplies the neutrons in a subcritical assembly that contains the waste to be transmuted. Unlike the LANL ATW, the proton target in Phoenix is composed of the MA waste itself. The neutrons have a hard spectrum, in contrast with the thermalized spectrum of the ATW. The Phoenix concept builds on existing oxide fuel technology developed under the FFTF program, together with aqueous separations technologies to reprocess both LWR and Phoenix spent fuel.

Since there is no plutonium in the Phoenix initial fuel charge, there is insufficient neutron generation for the MAs to burn both the radio-iodine and the technetium fission products. The discharged HLW containing technetium and the other fission products would be packaged for surface storage and, ultimately, geologic disposal. Thus, the scope of HLW transmutation would not be as extensive as in the ATW concept and is more like that of a fast spectrum, critical reactor approach, such as the ALMR.

The ATW and Phoenix concepts have several general features in common that can be contrasted with critical reactor-based concepts. Both laboratories point out that high fissile specific power can be attained that can result in fast burn-up rates of the actinides and certain fission products as well. BNL points out that the combination of high flux and hard spectrum in Phoenix results in shorter times to achieve given reductions in actinide inventories than possible with an ALMR.

Indeed, LANL believes that a neutron spallation source coupled to a fission blanket provides a fundamental enhancement in the effective number of neutrons per fission. Their calculations project a high-intensity thermal flux of 2 to 3 × 10^{15} neutrons/cm^2-s, which is about an order of magnitude higher than is typical of thermal power reactor systems (Bowman et al., 1991). For such a system, LANL projects two key advantages (Arthur, 1992b).

First, a high neutron flux could provide more efficient destruction of certain actinides by enabling a nucleus to undergo two sequential (n, α) reactions, increasing the probability of the nucleus capturing a second neutron before the state created by capture of the first neutron would decay. Thus, an ATW system could, in principle, efficiently transmute some actinides that normally cannot be transmuted well with a thermal neutron flux but require fast neutrons.

However, the high thermal flux of the ATW would lead to a build-up of higher MAs in the residual inventory as transmutation proceeds, in particular, ^{242}Cm, ^{244}Cm, ^{246}Cm, and ^{248}Cm as well as ^{252}Cf, all of which are significant neutron emitters. The level of radioactivity would be considerably higher than for the LWR and, of course, orders of magnitude higher than with a fast neutron spectrum. As discussed in Appendix F, the level of radioactivity may affect the quality assurance of the slurry/liquid target fuel during transmutation.

Second, the separation of accelerator and target/blanket assembly allows for rapid transmutation and entails relatively low inventories. Indeed, a main feature of the ATW, discussed in detail in Appendix F, is that it can transmute a given fraction of its TRU inventory far more rapidly than a fast reactor. This is a consequence of the lower critical concentrations characteristic of thermal reactors and the lower inventory in the on-line reprocessing system, as discussed later in this chapter.

Finally, both laboratories assert that an accelerator-based waste transmuter has potentially improved safety features vis-à-vis critical reactor concepts because, at least in theory, the assembly can be maintained subcritical, and shutdown can be accomplished rapidly by shutting off the accelerator. However, the discussion in Appendix F on the ATW and later in this chapter brings to light several types of transients that pose potentially significant safety issues for the accelerator-based concepts, in addition to the issues posed by decay-heat removal and target-heat dissipation.

Separations with Transmutation of Plutonium (only) [9]

An LWR or an ALMR would operate much more efficiently with ^{239}Pu as the recycle fuel than with recycle of all the TRU isotopes contained in the LWR spent fuel. This suggests a waste management concept intermediate between the once-through LWR fuel cycle and the full transmutation of all TRUs and selected fission products. That is, the accumulated LWR spent fuel could be reprocessed and only the separated plutonium recycled to either an LWR or ALMR. In addition, ^{14}C and ^{129}I could be captured, e.g., as off-gasses, and packaged in low-solubility waste forms that

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[9] The National Academy of Sciences had conducted another study on weapons plutonium and a report "Management and Disposition of Excess Weapons Plutonium" was published in 1994.

could be developed. The recovered uranium would be packaged for geologic disposal or possibly for recycle to an enrichment facility. An LWR could also transmute the long-lived ^{99}Tc and ^{129}I fission products if they were separated and recycled.

An ALMR could operate with the types of fuel previously developed for the fast breeder; the fast neutron spectrum would minimize higher plutonium build-up. With a LWR, the essential difference between this plutonium recycle concept and the "classic" thermal recycle of plutonium, demonstrated in the late 1960s, is that here the plutonium would be multiply recycled, resulting in a build-up of higher plutonium isotopes in the LWR thermal neutron spectrum. In particular, ^{240}Pu and ^{242}Pu are significant absorbers of thermal neutrons and reduce reactivity. Reactivity of the multiple recycle LWR fuel, however, can be maintained satisfactorily with make-up ^{239}Pu or uranium of moderate enrichment.

For such plutonium-only transmutation, present-day separations technology using PUREX may be sufficient, having process losses on the order of 1% for plutonium (see Chapter 3). The other TRUs would go to the HLW for geologic disposal, although some of the neptunium might also be recycled if that proved easy to do. The PUREX process waste might be contained in a glass waste form, as planned for the defense HLW. As noted above, a waste form would need to be developed for the ^{14}C. By also separating certain long-lived fission products, one could possibly devise other improved waste forms, e.g., for ^{99}Tc and ^{135} Cs. If pyroprocessing were used with an ALMR plutonium burner, additional new waste forms would be required. Of course, the big costs are those of LWR spent-fuel reprocessing and the all-remote fabrication of plutonium-bearing recycle fuel. Moreover, this waste management concept opens the door to the major public policy issues entailed in the commercial use of plutonium, in particular the relationship to U.S. nonproliferation policy (see Chapter 6).

The main benefits to waste management would come from (1) the improved waste forms and/or transmutation of the long-lived fission products and (2) waste form for ^{14}C, which is an apparent issue at present for the special case of the Yucca Mountain repository. However, it is problematic whether plutonium-only transmutation could capture other prospective benefits to waste management claimed for the full transmutation of the TRUs. First, transmuting the plutonium could reduce somewhat the calculated repository hazards from human intrusion scenarios in which waste is brought directly to the surface. The effect would depend on the time after repository closure at which the human intrusion event occurred.

For postclosure times of a hundred years or so, ^{238}Pu with an 87.7-year half-life is an important contributor to the TRU activity; this radionuclide would be transmuted. However, for postclosure times to several thousand years, the untransmuted ^{241}Am with an 433-year half-life would dominate the TRU activity. This would be especially true for the older spent fuel in which the ^{241}Pu had already decayed with a 14.4-year half-life to ^{241}Am. For postclosure times of 10,000 years or more, ^{239}Pu and ^{240}Pu would dominate the TRU activity and would be transmuted. However, when the ^{239}Pu is eliminated by transmutation, or has decayed, ^{237} Np would dominate the TRU activity.

In addition, plutonium is quite insoluble in groundwater under planned repository conditions. Thus, plutonium transmutation would have little direct effect on the dissolution-and-migration scenarios important for long-term repository risk (except by eliminating one of the precursors of ^{231}Pa). Also, removal of the plutonium would reduce the long-term heat load on the repository, although the untransmuted ^{241}Am would still be a major contributor. For the Yucca Mountain repository, however, decay heat is being considered as a means of keeping the waste containers dry while increasing the capacity of the repository, so that transmuting the plutonium (or other TRUs) may actually be a disadvantage for that site.

REDUCTION OF TRANSURANIC INVENTORIES

Introduction

This section examines the extent to which the amounts of TRUs in wastes from the various proposed transmuters can be made significantly smaller than the amount of TRUs in spent fuel in the reference once-through LWR fuel cycle. Inventories of TRUs in wastes from fuel reprocessing and TRU recycle are considered, together with inventories of untransmuted TRUs in the transmuter and in the associated facilities for reprocessing and fabrication of recycled material. To defray the cost of transmutation, all of the transmutation concepts propose generating and selling electrical energy resulting from the fission of TRUs. In this section the inventories of TRUs in the transmutation fuel cycle and in the reference once-through fuel cycle are compared for the same amount of electricity generated.

Typically, only a small fraction of the TRUs in a transmuter would be transmuted while exposed to reactor neutrons during an irradiation cycle. The fuel discharged from the reactor would be reprocessed to remove fission products and the recovered TRUs would be recycled. Special techniques for reprocessing are specified in order to reduce the amount of TRUs lost to the reprocessing wastes. Process-loss fractions of the order of 0.001 to 0.0001[10] or less have

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[10] ANL is said to be developing pyrochemical separations with process-loss fractions of 0.001 to 0.0001 for each processing cycle. Developers of the TRUEX aqueous separation, to be used in conjunction with PUREX aqueous reprocessing, expect that process-loss fractions of 0.001 to 0.0001 can be achieved. LANL, the developer of the ATW, expects process-loss fractions of 0.0002 or better for neptunium and plutonium and to 0.000003 or better for americium and curium, sufficient so that the resulting TRU waste can be treated as low-level waste (Arthur, 1993).

been proposed. However, TRUs must be recycled many times through the reactor and its external fuel cycle before being transmuted, so the actual fraction lost to the reprocessing waste is greater than the fraction lost in each reprocessing cycle. Also, for safety and fuel-cycle economics, several of the transmutation concepts convert fertile ^{238}U or ^{232}Th to additional TRUs that must also be recycled and transmuted. Consequently, even with new separations that would reduce the losses to waste to 0.001 to 0.00001 of the amount processed, the inventories of TRUs in the waste would be far greater than what might be expected by multiplying the TRU inventories in LWR spent fuel by the process-loss fraction.

For many of the transmuter concepts, such as the ALMR, the inventories of TRUs in the reactor and its external reprocessing-refabrication fuel cycle are large. In some transmuter concepts much of the TRUs supplied to the transmuter during its life would remain in the reactor and its fuel cycle. This residual inventory must be accounted for as potential high-level waste. Consequently, as long as nuclear power from transmuters is to be continued, the TRU inventory at the end of life of each transmuter would be transferred to a succeeding transmuter. To achieve the transmutation goals, it would be necessary to operate the transmuters long enough so that the above-ground inventory in the transmuter and its fuel cycle is no longer much larger in magnitude than the inventory in the total waste produced. Therefore, the time-dependent inventories of TRUs in the transmuter and its fuel cycle should be included when analyzing the extent to which transmutation can reduce TRU inventories below the inventories in the reference once-through LWR fuel cycle.

For a given number of first-generation transmuters, constant electric power from transmuters would result if each succeeding transmuter were of the same electric power as the first-generation transmuter. Alternatively, each generation of follow-on nonbreeding transmuters[11] could be of smaller electrical power as the total TRU inventory is consumed, resulting in the phase-out of nuclear power at a rate consistent with the necessary reduction in total TRUs that will become radioactive waste. For nonbreeding transmuters, additional LWRs could eventually be required to furnish make-up TRUs to fuel the transmuters, once the stockpile of stored LWR fuel has been exhausted.

Calculation for the various transmutation concepts of the time-dependent ratio of the amount of TRUs in waste from the reference LWR once-through fuel cycle to the amount of TRUs in the transmutation fuel cycle is the subject of this section.

The Transuranic (TRU) Ratio

The following analysis (Hebel et al., 1978; Pigford and Choi, 1991) describes the extent of TRU inventory reduction for the various transmuter concepts as a function of time. It is important to use an index that measures the extent to which TRU inventories in the transmuter, in its waste, and in its fuel cycle would be smaller than the inventory of TRUs for the reference fuel cycle of once-through LWRs. This index is called the "transuranic (TRU) ratio" $\psi(t)$, defined as

$$\psi(t) = \frac{\text{total inventory of transuranics sent to waste disposal in time for the reference once-through LWR fuel cycle, if no fuel reprocessing, no recycle, and no transmutation}}{\text{total inventory of transuranics at time } t \text{ in the transmuter, in its fuel cycle, and in process wastes}}$$

TRUs supplied to the transmuter system for start-up and for make-up fuel can be obtained by reprocessing existing LWR spent fuel and/or by reprocessing spent fuel from future LWRs. In calculating quantities for the equivalent reference fuel cycle, i.e., for the numerator of $\psi(t)$, that same amount of TRUs must be assumed to go directly to waste disposal. Also, in the trnasmutation fuel cycle, transmuters generate a specified electric power $P(t)$ and a specified total electrical energy by time t. To maintain equivalence, the TRU inventory in the nontransmutation fuel cycle considered in the numerator of $\psi(t)$ must also include the TRUs that would be produced by reference LWRs producing the same electric power $P(t)$ and the same electrical energy as the transmuters. All of these TRUs calculated for the non-transmutation fuel cycle are assumed to be sent to waste disposal.

A more direct measure of the extent of depletion of TRUs by transmutation is the ratio of the total amount of TRUs supplied to the transmuter to the inventory of TRUs in the transmuter system and in its waste. This ratio is defined as the "depletion ratio" $\chi(t)$. It is lower in magnitude than the TRU ratio $\psi(t)$. To be meaningful, however, the comparison of waste-disposal benefits for alternative electric power systems must be made on the basis of the same electric power and the same electric energy. Therefore, the transmutation concepts should have the benefit incorporated into the TRU ratio $\psi(t)$.

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[11] A nonbreeding transmutor has a breeding ratio less than unity.

Scenarios for Constant Transmuter Power

Transmutation devices could replace some or all of the LWRs (or other reactors with a once-through fuel cycle) that provide continuous generation of electricity. A given initial amount of TRUs, such as the lifetime inventory from all current LWRs, could be used as the start-up inventory of "breeder" transmutation devices (i.e., breeding ratio of unity or greater). As an alternative, "nonbreeder" transmutation devices (i.e., breeding ratios less than unity) could be used for more rapid reduction of the TRU inventory, as suggested by DOE/NE (Young and U.S. Department of Energy, 1991). The nonbreeders would require make-up fissile material for ongoing operation, as well as start-up inventory.

A scenario of steady transmuter power has many options for deploying transmuters. Three options are treated in this subsection, corresponding to (1) continuous operation of breeder transmuters; (2) initial operation of transmuters as nonbreeders, later switching to breeders; and (3) continuous operation of nonbreeder transmuters and TRU-producing reactors, such as LWRs. Each of these options could involve an initial period in which the transmuters are started and fueled with TRUs recovered from the stockpile of LWR spent fuel. The desired goal would be to reduce the total TRU inventory well below that of the reference once-through LWR fuel cycle. The extent of that reduction would increase with time, until the option attains an asymptotic state in which the inventory of TRUs in the accumulated reprocessing waste becomes much larger than the inventory in the transmuter and its fuel cycle.

1. Breeder transmutation devices could be started utilizing the inventory of TRUs accumulated from reprocessing existing spent LWR fuel for breeder start-up. No new LWRs would be required. Even though the TRU inventory within the reactor and fuel cycle would remain constant with time, together with the inventory in reprocessing waste, it would be less than the inventory in unreprocessed fuel from the once-through fuel cycle if it delivered the same amount of electrical energy.
2. The new transmutation power plants could be operated as nonbreeders until all the accumulated inventory of TRUs from reprocessing existing LWR spent fuel has been utilized for start-up and refueling. These transmutation systems could then be converted to breeders. No new LWRs would be required. The ultimate degree of inventory reduction would be the same as that obtained in option (1).
3. The new transmutation power plants could be operated as nonbreeders. TRUs from the stockpiled LWR spent fuel would be used to start the first transmuters and to refuel these and subsequent transmuters. If the stockpile of spent LWR fuel is not sufficiently large, LWRs (or some equivalent nuclear power plant with a once-through fuel cycle) could be operated to supply make-up fissile material for the later-generation transmuters.

Declining Power Scenario

Nonbreeding transmutation devices could be constructed and operated to transmute as much as possible of a given initial inventory of TRUs. Each transmutation device, operating at a given thermal power, would require a certain inventory of TRUs in its critical or subcritical reactor and in its fuel cycle. If the desired inventory reduction is greater than can be achieved during the life of a given initial number of transmutation devices, the inventory remaining at the end of life of the first-generation transmuters can be used to start and fuel a smaller number of second-generation transmuters. In each subsequent generation there would be less total inventory, so that the total thermal power of the transmuters—i.e., the number of transmuters of a given thermal power in the system—would decrease with each generation until, finally, there would not be enough inventory and reactivity to operate a single transmuter module. This process would be realistic if it were planned to discontinue nuclear power after a desired inventory reduction was achieved.

Inventories and Transmutation Rates

The following results are presented for the TRU ratio (t) for the many transmutation options and their variations described in detail in various appendices to this report. The numerical parameters used in the calculations are summarized in Table 4-3. The transmuters treated here are the following: (1) the ALMR/IFR for several values of the breeding ratio; (2) four ATW concepts, including the baseline aqueous system (ATW-1) and three advanced nonaqueous systems (ATW-2, ATW-3, and ATW-4); (3) the LWR with full TRU recycle; and (4) the PBR TRU burner. As discussed in the ALMR section of this report, there is not a uniform progression of inventories (I), specific burn-up rate (B/I), and other properties with decreasing breeding ratio. This is because some of the ALMR designs are the modular PRISM concept, others are more nearly homogenous cores, and one (BR = 0.22) specifies refueling the entire reactor core at the end of an irradiation cycle. Some differences in the six ATW concepts are explained in the table footnotes.

These data are used to illustrate the extent of long-term transmutation by ALMRs, for both steady and declining power. Similar calculations for other transmuters are presented to point out the differences in TRU transmutation by the various concepts.

A recent publication by J. C. Lee and J. Du (1994) suggests that the calculations for a LWR TRU burner may be optimistic. If first-generation LWR TRU transmuters are fueled with TRUs recovered from stored and aged fuel discharged from once-through LWRs, the decay of ^{241}Pu to ^{241}Am during decades of storage of the once-through discharge fuel should be considered. This was not included in the calculations by Gorrell (1979) that were the basis for the data on LWR TRU transmuters in Table 4-2. Instead, Gorrell assumed recycle of actinides from contemporaneous LWRs, assuming two years from fuel discharge to recycle as MOX fuel.

Decades of storage prior to recycle could result in decay of most of the ^{241}Pu in fuel at the time of reactor discharge. The decay product, ^{241}Am, is thermally nonfissile and has a high cross section for absorbing thermal neutrons. Thus, the highly fissile ^{241}Pu that would aid criticality in lessaged recycled TRUs would have been replaced by a strong neutron absorber. Consequently, greater concentrations of fissile actinides would have to be present in the MOX fuel for criticality. The inventory of TRUs would increase, and it would take longer to reach a near-equilibrium fuel-cycle inventory. Also, the calculations by Lee and Du indicate a lower net burn-up rate of TRUs than that calculated in Gorrell's data.

Lee and Du also assume that all uranium recovered from reprocessing discharged LWR fuel would also be recycled directly as MOX fuel. However, uranium recycle directly as MOX fuel in LWRs is not contemplated by those countries that are pursuing the option of fuel reprocessing. Instead, the recovered uranium would be recycled to a facility for uranium isotopic enrichment, or it could be stored. Recycling uranium directly to the reactor would increase the concentrations of neutron-absorbing ^{236}U and of the neutron-capture products ^{237}Np and ^{238}Pu, all of which absorb thermal neutrons. For the LWR-plutonium and LWR-TRU transmuters considered in the present report, the recovered uranium would not be recycled directly as MOX fuel, regardless of the age of the fuel to be reprocessed for TRU recycle. Calculations for this fuel cycle are based on data of Gorrell, who did not assume recycle of uranium directly as MOX fuel. Therefore, the calculations by Lee and Du have overestimated the extent of neutron absorption from ^{236} U, ^{237}Np, and ^{238}Pu.

The calculations by Lee and Du indicate less net destruction of TRUs than do the calculations by Gorrell. Whether this would still be true for the LWR-TRU fuel cycle, without direct uranium recycle as MOX fuel, remains to be determined from further calculations.

If LWR TRU transmuters were to operate for the very long times considered in this study, the effects of fueling first-generation transmuters with TRUs from long-cooled LWR discharge must ultimately disappear. For example, in the following section, *Constant-Power LWR Transmuters* , times of the order of thousands of years to achieve a significant net reduction in total TRU inventory are calculated. The calculations by Gorrell would then describe the fuelcycle properties that would ultimately emerge. The inventories and burn-up rates calculated by Lee and Du, if corrected for no direct recycle of uranium, would better describe the characteristics of transmuters during the first few reactor generations.

Thus, the issues resulting from the recent calculations by Lee and Du cannot be resolved without further calculations. Such calculations would be lengthy and are not within the scope and schedule of the present study. Our illustrations herein of the possible features of LWR plutonium and LWR TRU transmuters are based necessarily on data from Gorrell's calculations.

Constant-Power ALMRs, Unlimited Supply of TRUs From Stockpile of LWR Spent Fuel

The TRU inventories for ALMR burners of 0.65 breeding ratio for the first 100 years for the simple case of constant power and an unlimited stockpile of spent fuel containing TRUs are shown in Figure 4-1. The scenario is conceptually simple and is presented to aid in understanding. Also, the resulting TRU ratio is identical to that of more realistic scenarios of a limited LWR-spent-fuel stockpile, as illustrated later in Figure 4-3. The total electrical power assumed to calculate inventories for Figure 4-1 is 30.4 GWe, to facilitate comparison with Figure 4-3. However, the value of the TRU ratio (t) is independent of the power level. In this figure it is assumed that at end of life, each ALMR is replaced with an identical ALMR that receives the reactor and fuel-cycle inventory of TRUs from its predecessor. Calculations during the first 10 years are only approximate because steady-state fuel-cycle quantities are assumed.

Assuming, for simplicity, that all initial ALMRs begin operating at time zero, the total TRU inventory in the ALMRs and their fuel cycles is 305 Mg and is essentially constant with time. For a process loss to waste of only 0.1% per reprocessing cycle, the TRUs appearing in waste during the first 100 years would be relatively small, only about 8.3 Mg. The total TRU inventory for the ALMR option is given by line no. 2.

If there were no ALMR transmuters, the stockpile of spent LWR fuel withdrawn for reprocessing to start and fuel ALMRs would instead go directly to waste disposal. That amount is shown by the dashed line no. 3 of Figure 4-1, labeled "TRUs used by ALMRs." Further, with no ALMRs the reference once-through LWRs would produce the same constant electrical power as did the ALMRs. At any instant the inventory of TRUs in the spent fuel from those reference LWRs is the difference between line no. 1, labeled "Total direct disposal," and the dashed line.

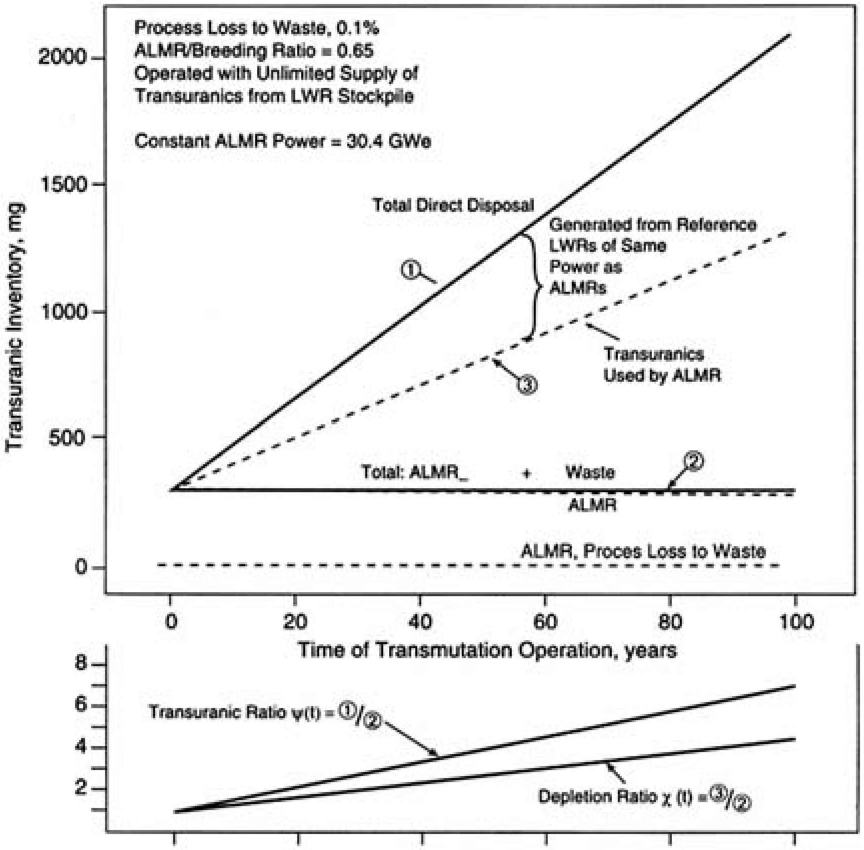

FIGURE 4-1 TRU inventory, TRU ratio and depletion ratio versus time for constant-power ALMR, unlimited stockpile of LWR spent fuel.

SOURCE: Choi and Pigford, 1994.

The ratio of the inventory of line no. 1 to the inventory of line no. 2 is the TRU ratio (*t*), shown on the lower plot of Figure 4-1 for the same time scale. The TRU ratio reaches a value of 6.9 at 100 years. Reducing the TRU inventory by only a factor of 6.9 below that of the reference once-through fuel cycle is far from the goals proposed for transmutation, yet that would be the reduction if ALMRs were operated at constant power for 100 years and then terminated.

Line no. 3 of Figure 4-1 shows the total integrated quantity of TRUs supplied to the ALMR transmuters from time zero to time *t*. The ratio of line no. 3 to line no. 2 is the depletion ratio (*t*), shown in the lower plot of Figure 4-1. It is the factor by which TRUs supplied to the transmuter are depleted by transmutation. However, the depletion ratio does not reflect the additional benefit to the transmutation system that results from the reference once-through LWR reactors of the same electrical power producing additional TRUs that would also be sent to waste disposal. Therefore, it is the TRU ratio (*t*) that is emphasized throughout this chapter.

More extensive reduction below that of the reference fuel cycle would require much longer operation, as shown in Figure 4-2. Here the TRU ratios are plotted as a function of operating time for ALMRs of various breeding ratios (BR) and for two different values (0.001 and 0.0001) of the process-loss fraction. These curves also apply to other scenarios involving constant power of transmuters, such as simultaneous operation of LWRs to supply make-up TRUs to nonbreeding ALMRs, as discussed in the next section. The curves of Figure 4-2 are calculated to times that are probably unrealistic for any but the breeding ALMRs (BR = 0.96 and 1.1). Uranium resource[12] limitations may prelude simultaneous operation of LWRs for such long times.

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[12] Recent calculations by ORNL, referred to in the ALMR chapter, suggest that if the cost of reprocessing LWR spent fuel for TRU recovery is as high as $1,000/kg, the LWR fuel cycle could be more economical than that of an ALMR even by using natural uranium from sea water, if uranium could be obtained from sea water by a new Japanese process for about $150/lb. If so, this would considerably extend the era for competitiveness of the LWR once-through fuel cycle compared with the ALMR fuel cycle that uses TRUs from spent LWR fuel.

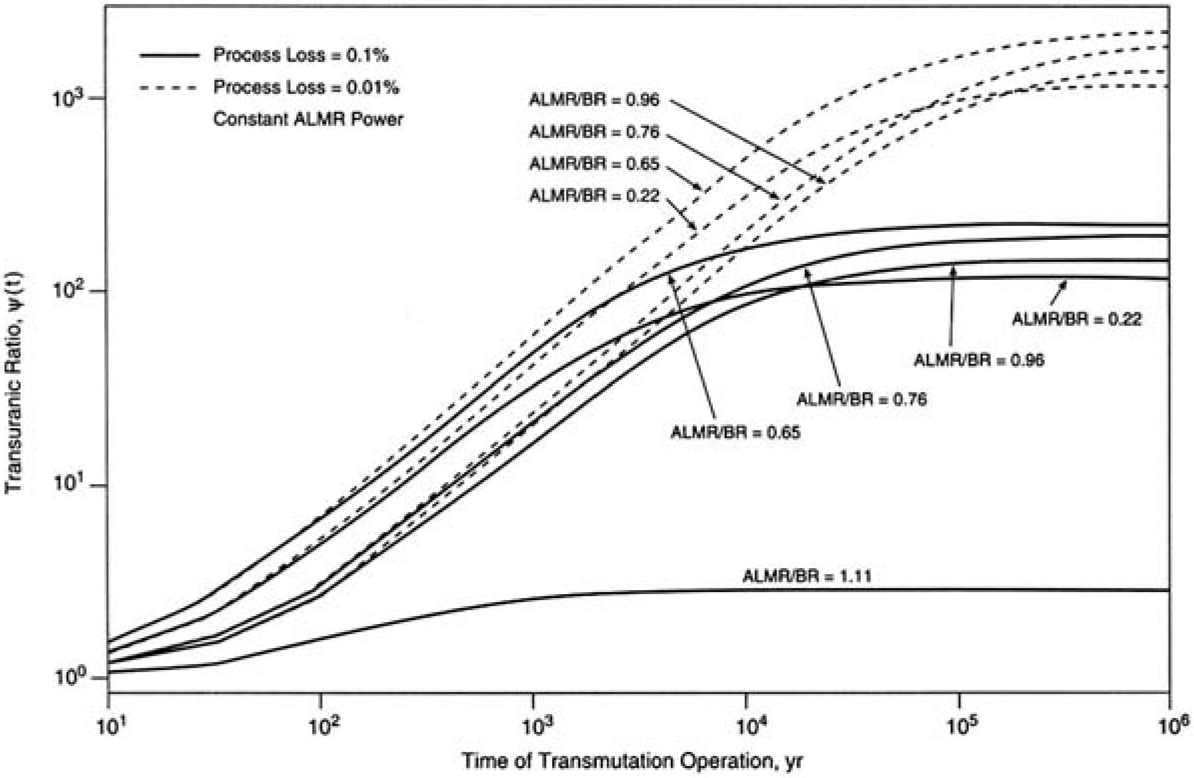

FIGURE 4-2 TRU ratio and depletion ratio versus time for ALMRs at constant power, effect of process loss.
SOURCE: Choi, J. S., and T. H. Pigford. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

Figure 4-2 shows that for a given breeding ratio the TRU ratio approaches asymptotically a constant value. The values of this asymptotic ratio are given in Table 4-2. For each breeding ratio the constant-power time constant τ, given as the time for the TRU ratio to reach within a factor of (1 – 1/e) of the asymptotic value, is also listed in Table 4-2. The time constant is about 6,000 years for 0.65 breeding ratio and about 14,000 for a break-even breeder.[13] The predicted TRU ratios at 100 years are listed in Table 4-3.

Both Figure 4-2 and Table 4-3 show that the attainable TRU ratio is affected little by the processing-loss fraction during the first few hundred years. Reducing the processing-loss fraction can affect the reduction factor appreciably only after many thousands of years.

Assuming that nuclear power is to continue in the future at a steady power level, and assuming that transmutation by ALMRs is desired to benefit ultimate waste by reducing TRU inventories by even as little as an order of magnitude, Figures 4-2 and 4-3 show that a commitment would have to be made to continue ALMRs and their progeny for many centuries.

Constant-Power ALMRs, Limited Stockpile of LWR Spent Fuel, Additional LWRs

Figure 4-3 shows the time-dependent TRU inventories for the mixed ALMR-LWR fuel cycle. This is the fuel cycle proposed by DOE, except that here it is assumed that all first-generation ALMRs start operating simultaneously. The 612 Mg of TRUs that can be obtained by reprocessing the 62,000 Mg stockpile of LWR spent fuel is reduced at time zero when 305 Mg is withdrawn to start the ALMRs. The remaining 307 Mg is used as make-up fuel for the first-generation ALMRs during their operating life. Thereafter, 39.9 GWe of new LWRs begin operation to furnish make-up TRUs for the second-generation and later-generation

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[13] According to the ALMR project, an ALMR with a breeding ratio of 0.96 is actually a "break-even" breeder. This anomaly results from the arbitrary definition of breeding ratio adopted by the project. The project defines "breeding ratio" as the rate of production of ^{239}Pu and ^{241}Pu by neutron absorption in fertile species divided by the rate of destruction of ^{239}Pu and ^{241}Pu by neutron fission and by nonfission capture of neutrons. The definition is arbitrary, because all TRUs fission in the fast neutron spectrum of an ALMR.

ALMRs. For the reference fuel cycle the total electric power is 70.3 GWe, the sum of the electrical power for the ALMRs and LWRs in the transmutation fuel cycle. The total inventory of TRUs to waste disposal for this mixed fuel cycle is identical to that in Figure 4-1, and the TRU ratios for the two scenarios (Figures 4-1 and 4-3) are identical. The long-term TRU ratios shown in Figure 4-2, and the discussion thereof, also apply here.

TABLE 4-2 Numerical Parameters Used in the Calculations for Inventory Reduction Factors for ALRMs, LWRs, ATWs, and PBR

Transmutors[a]	I[b] (kg)	Cycle[c] (yr)	F/I[d] (yr^{-1})	D/I[e] (yr^{-1})	B/I[f] (yr^{-1})	P/I[g] (yr^{-1})	$_{inf}$[h]	τ[i] (yr)
ALMRs								
β=1.1 1[j]	27,200	1.88	0.100	0.104	-.00424	0.0132	3	190
0.98[j]	19,400	1.61	0.121	0.121	0	0.0185	153	14,100
0.76[k]	27,600	1.88	0.108	0.099	0.00904	0.0130	204	15,800
0.65[j]	14,000	1.67	0.262	0.229	0.0336	0.0256	220	6,070
0.62[k]	14,400	1.16	0.175	0.149	0.0259	0.0249	290	9,800
0.22[l]	34,900	0.56	0.347	0.316	0.0313	0.0103	120	4,800
LWR, Pu[m]	17,200	1.00	0.200	0.152	0.0481	0.0208	345	8,560
LWR, TRU[n]	21,600	1.00	0.200	0.162	0.0377	0.0166	272	8,530
ATWs								
Aqueous[o]	15,550	0.146	7.00	6.85	0.1458	0.0231	24	230
Nonaq[p]	600	0.083	14.1	12.0	2.05	0.598	188	120
Nonaq/Th 75%, Pa[p]	480	0.083	12.7	12.0	0.698	0.748	113	130
Nonaq/Th 75%[p]	1,130	0.083	12.3	12.0	0.297	0.318	50	135
Nonaq/Th 100%, Pa[p]	340	0.083	12.0	12.0	0	1.06	88	110
Nonaq/Th 100% [p]	1,280	0.083	12.0	12.0	0	0.280	23	130
PBR[q]	1,425	—	1.46	0.615	0.845	0.176	700	1,200

[a] β is the breeding ratio, defined as the ratio of thermally fissile ^{233}U, ^{234}U, ^{239}Pu, ^{241}Pu production to thermally fissile destruction.

[b] Inventory of TRUs in transmutor and fuel cycle at steady state, scaled to the power level of 1,395 MWe.

[c] For the solid-fuel ALMRs and LWR the cycle time is the chronological time between refuelings. For the fluid-fuel ATWs the cycle time is the time to process the blanket inventory. All times are for a capacity factor of 0.8.

[d] F is the refueling rate of TRUs.

[e] D is the discharge rate of TRUs from the core and blanket.

[f] B is the rate that TRUs must be supplied from an external source. At steady state, F = D + B.

[g] P is the rate of production of TRUs from a 1,395 MWe PWR, 33 MWd/kg, 0.80 capacity factor, equal to 359 kg/yr (Benedict et al., 1981; D.O.E., 1987).

[h] $_{inf}$ is the asymptotic reduction factor, for the process-loss fraction of 0.001.

[i] τ is the time to reach (1 - 1/e) of the asymptotic value, for $\gamma = 1,000$.

[j] Data from General Electric for a PRISM ALMR with core and blanket. Includes inventory in 2-year external cycle for cooling and reprocessing discharged ALMR fuel (M.W. Thompson, private communication, 1991; K. Wu, private communication, 1991).

[k] Data from General Electric for a PRISM ALMR with a homogenous core and no blanket. Includes inventory in 2-year external cycle for cooling and reprocessing discharged ALMR fuel. (M.W. Thompson, private communication, 1991).

[l] Derived from ANL data for a 450 MWe ALMR with no blanket and core charged entirely with TRUs, scaled to 1,395 MWe and 80 percent capacity factor (Johnson et al., 1990). Includes inventory in 2-year external cycle for cooling and reprocessing discharged ALMR fuel.

[m] From Pigford and Yang (1977). See also Hebel et al. (1978: Figure 8, p. 57). Based on data for total plutonium. Only plutonium is recycled.

[n] Derived from data of Gorrell (1979).

[o] Derived from equations and data given by Davidson (1992).

[p] Derived from data of Bowman (C.D. Bowman, private communication, 1992). The notation "nonaqueous/Th, 75%, Pa" indicates that the fluid fuel is nonaqueous, ^{232}Th is added as a fertile material to breed ^{233}Pa and ^{233}U, 75% of the thermal energy results from fissioning bred uranium, and protactinium is separated rapidly in the on-line coupled reprocessing system. For the systems using ^{232}Th, computation of "TRUs" also includes the protactinium and uranium.

[q] Derived from data of Kang and Kazimi (1993).

TABLE 4-3 TRU Ratios for Constant-Power ALMRs at 100 Years

TRU Ratio (t)		
Breeding Ratio	0.001 Loss Fraction	0.0001 Loss Fraction
0.22[a]	4.98	5.14
0.65	6.74	6.90
0.96	2.81	2.85

[a] The ALMR with 0.22 breeding ratio contains negligible quantities of ^{238}U, the usual fertile material to breed TRUs. In that sense, it does not breed from natural fertile material. The value of 0.22 results from the ALMR project's arbitrary definition of breeding ratio, i.e., it treats neutron absorption in ^{238}Pu and ^{240}Pu as "breeding."

Declining-Power ALMRs

Figure 4-4 shows the time-dependent inventories and TRU ratio for a declining-power scenario, chosen to achieve

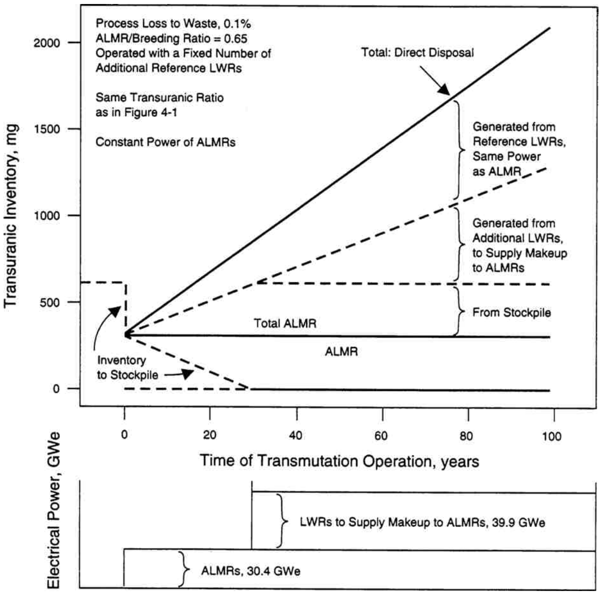

FIGURE 4-3 TRU inventory and ratio versus time for limited stockpile of LWR spent fuel, constant-power ALMR.
SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

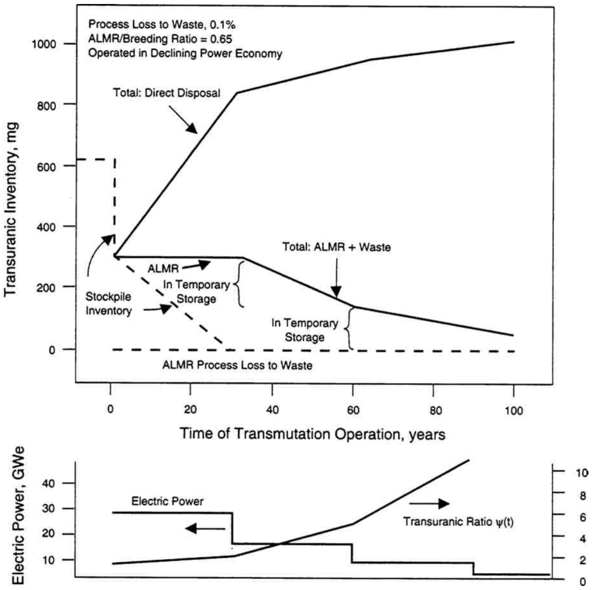

FIGURE 4-4 TRU inventory and ratio versus time for declining-power ALMRs.
SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

more rapid reduction in TRU inventories than is possible for the constant-power scenario. During the first 30 years, 30.4 GWe of ALMRs of 0.65 breeding ratio are assumed to operate, using the entire TRUs available from the 62,000 Mg stockpile of LWR spent fuel otherwise destined for the proposed Yucca Mountain repository. Rather than constructing additional LWRs to fuel the second-generation ALMRs, the inventory of TRUs from the first-generation ALMRs is committed to start and refuel 15.1 GWe of second-generation ALMRs. Similarly, only 7.5 GWe of third-generation ALMRs would be used, and so on, until only a single critical module of an ALMR is finally used. In this way the TRU ratio and the extent of TRU depletion increase more rapidly with time than with constant power. In 100 years a TRU ratio of 11 could be obtained, compared to only 6.9 for the constant-power scenario.

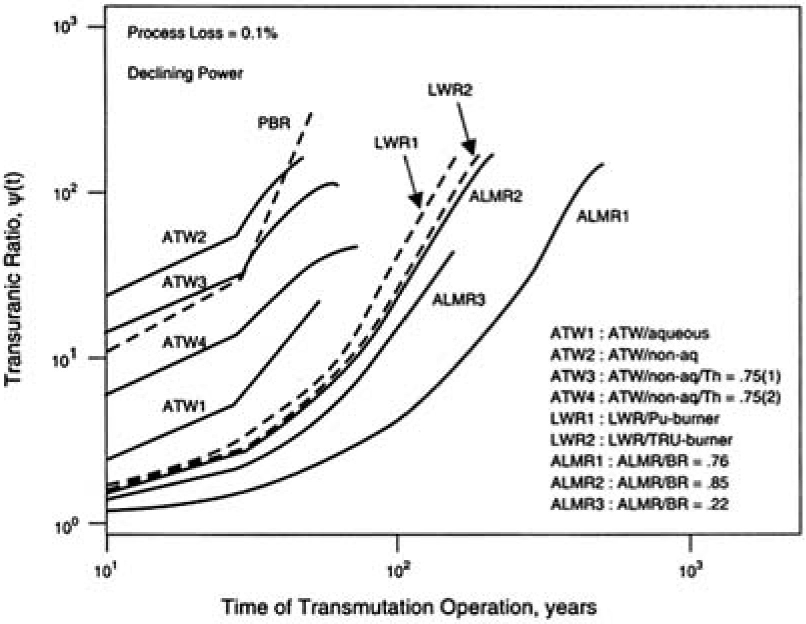

FIGURE 4-5 TRU ratios versus time for declining-power ATWs, LWRs, and ALMRs.
SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

In the declining-power ALMRs illustrated in Figure 4-4 the actual inventory of TRUs in the ALMRs would be reduced by only a factor of 8.1 in 100 years. Thus, the depletion ratio (*t*) would be 8.1, as compared to the TRU ratio (*t*) of 11 in 100 years. However, as explained earlier, actual depletion is not the proper figure of merit. The ALMRs must be credited with generating electrical energy during the depletion period. In the reference scenario of LWRs of the same electrical energy, additional TRUs would be produced, in addition to the original 612 Mg present in the LWR spent-fuel stockpile. Therefore, the proper index to illustrate the effectiveness of ALMRs in depleting TRUs is the TRU ratio (*t*), as adopted throughout this analysis.

The longer-term TRU ratios for ALMRs, ATWs, and LWRs in a declining-power economy are shown in Figure 4-5. The curves terminate when the remaining TRU inventory is not sufficient to fuel a critical module of an ALMR. For the ALMR of 0.65 breeding ratio, a TRU ratio of 100 could be obtained in about 200 years. This illustrates again the long-term commitment, even for a declining-power scenario, that would be required to achieve inventory reductions that are proposed as significant for waste disposal.

Curves in Figure 4-5 for other transmuters are discussed later in the chapter.

Constant-Power ATWs

TRU inventories for constant-power aqueous ATWs, with a total saleable electrical power of 10.2 GWe, are shown in Figure 4-6. They are about threefold less than for the ALMR scenarios. This is a consequence of the aqueous ATW's lower net thermal efficiency. The total saleable electrical power from the aqueous ATW needed to utilize the 62,000-Mg stockpile of LWR spent fuel is lower than for ALMRs because of the relatively low net thermal efficiency of the aqueous ATW.[14] The aqueous ATW's saleable electric power is also lower than that of the ALMR of

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[14] The low net thermal efficiency of the aqueous ATW is a consequence of the limited system pressure and fuel-coolant temperature of the calandria-type pressure-tube reactor lattice and the need to supply electricity to the ATW's accelerator, as explained in the ATW section of this report.

0.65 breeding ratio (cf) (see Figure 4-3) because the aqueous ATW contains no fertile ^{238}U or ^{232}Th that would breed new TRUs. Since transmutation of TRUs is mainly by fission, their transmutation at a given total rate requires essentially the same thermal power for any transmutation system[15] . However, for no internal breeding of new TRUs, as in the aqueous ATW, all of the thermal power is devoted to transmuting TRUs from LWRs. Thus, if the aqueous ATW were required to transmute LWR transuranics at the same rate as an ALMR of 0.65 breeding ratio, less thermal power would be required. In addition, because of the aqueous ATW's low net thermal efficiency, far less saleable electrical energy would be produced by the aqueous ATW.

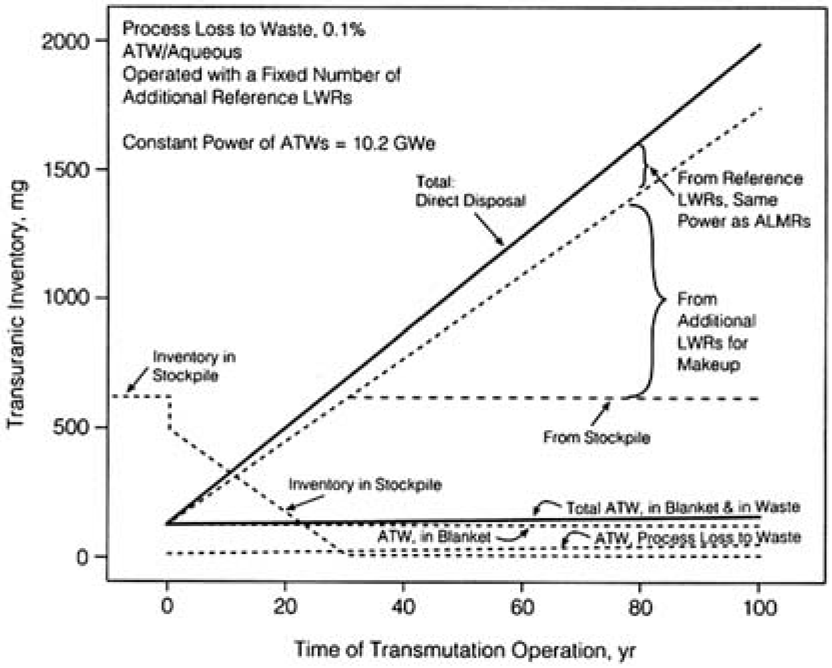

FIGURE 4-6 TRU inventory versus time for constant-power aqueous ATW, limited stockpile of LWR spent fuel. SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

Actually, the aqueous ATW's system inventory of TRUs is much lower than that of an ALMR for a given thermal power. This is a result of the lower fissile inventory needed for criticality in the heavy-water-moderated thermal-neutron spectrum of the ATW, the very high fissile specific power (thermal power per unit mass of fissile TRUs) of the ATW's slurry fuel, and the more rapid reprocessing and smaller external inventory of the ATW's coupled on-line reprocessing. As a result, less of the stockpile inventory is needed to start the ATWs, as seen by comparing Figures 4-3 and 4-6. Thus, an even greater transmutation rate, and hence greater thermal power, is needed for the aqueous ATW to utilize the remaining stockpile inventory of TRUs during the first generation of transmuters. Even so, the ATW's lower net thermal efficiency and lower breeding ratio result in the much lower saleable electrical power than from the ALMR of Figure 4-1. Whereas low net thermal efficiency results in greater transmutation rate per unit of net electrical power, it does not favor overall economics if the bulk of the transmutation cost is expected to be paid for by the sale of electricity.

For the same fractional loss per processing cycle, the amount of TRUs lost to reprocessing waste is greater for the ATW than for the ALMR because of the more frequent processing of the entire ATW inventory through the ATW's closely coupled reprocessing system. However, during the first few hundred years the inventory of TRUs in accumulated waste is less than the inventory in the transmuter system.

Time-dependent TRU ratios for constant-power ATWs, both aqueous and nonaqueous and effect of process losses, are shown as a function of time in Figure 4-7. Data in Table 4-2 show that the ratio B/I of the transmutation rate of LWR TRUs to the TRU inventory is much higher for the nonaqueous ATWs than for the aqueous ATW, a conse

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[15] Some additional transmutation of TRUs results from neutron capture and decay, especially in thermal reactors.

quence of the higher fissile-specific power[16] in the nonaqueous ATW cores. Even the nonaqueous ATW with 75% of the fissions from internal breeding from ^{232}Th achieves more rapid net burn-up of LWR TRUs, per unit system inventory, than does the aqueous ATW. The TRU ratios for the nonaqueous ATWs are higher. Because of the high specific powers of all the ATWs, the TRU ratios rise much more rapidly than for ALMRs. Time constants of only a few hundred years for the ATWs are listed in Table 4-2. However, the ATWs' asymptotic TRU ratios are, in many instances, lower than those of the ALMRs, because of the more frequent reprocessing in the ATWs. The TRU ratios attainable in 100 years with constant-power ATWs are shown in Table 4-4.

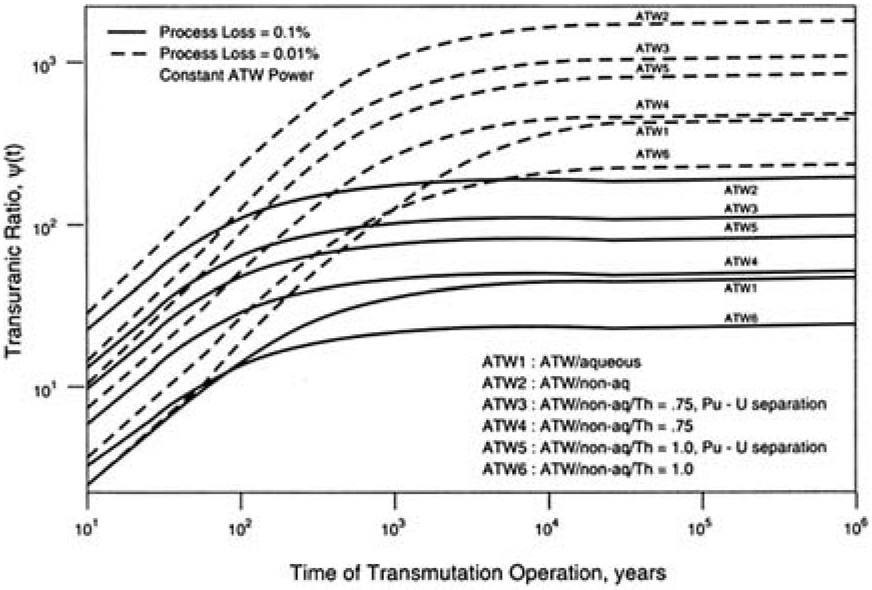

FIGURE 4-7 TRU ratio versus time for constant-power ATWs, effect of process loss. SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

Declining-Power ATWs

Figure 4-7 shows the ATW TRU ratios (*t*) for the constant power scenario and effect of process losses. Figure 4-8 shows the ATW TRU inventory for the ATW aqueous operating in a declining-power economy. For each ATW, the curve terminates when the inventory has reached that for the smallest module specified by LANL for its ATW designs. The ultimate TRU ratios are about the same as those obtainable from the ALMRs, but are achieved in less than 100 years.

TABLE 4-4 TRU Ratios for Constant-Power ATWs at 100 Years

TRU Ratio (*t*)		
ATW[a]	0.001 Loss Fraction	0.0001 Loss Fraction
Aqueous	13.1	17.3
Nonaqueous	110	233
Nonaq/Th=.75(1)	64.0	128
Nonaq/Th=.75(2)	28.0	55.6
Nonaq/Th=1.0(1)	48.3	95.1
Nonaq/Th=1.0(2)	13.1	25.9

[a] See Table 4-2 for explanation of the various ATW cases.

Constant-Power LWR Transmuters

Figure 4-9 shows the curves of the TRU ratio (*t*) for the LWR transmuters operating at constant power and effect of process losses. The TRU ratios are only slightly higher than those of the ALMR of 0.65 breeding ratio, and the time constants are nearly the same. An LWR operated only as a plutonium burner results in slightly higher TRU ratios than does an LWR in which all TRUs are recycled. These curves for the LWR burner would also apply to LWR reactors op

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[16] The fissile-specific power for the transmutation system is the ratio of the thermal power to the system inventory of fissile material. It is proportional to the product of the average neutron flux in the reactor core, the microscopic fission cross section, and the ratio of inventory in the reactor to the total system inventory. The flux-cross-section product is limited by the cooling rate.

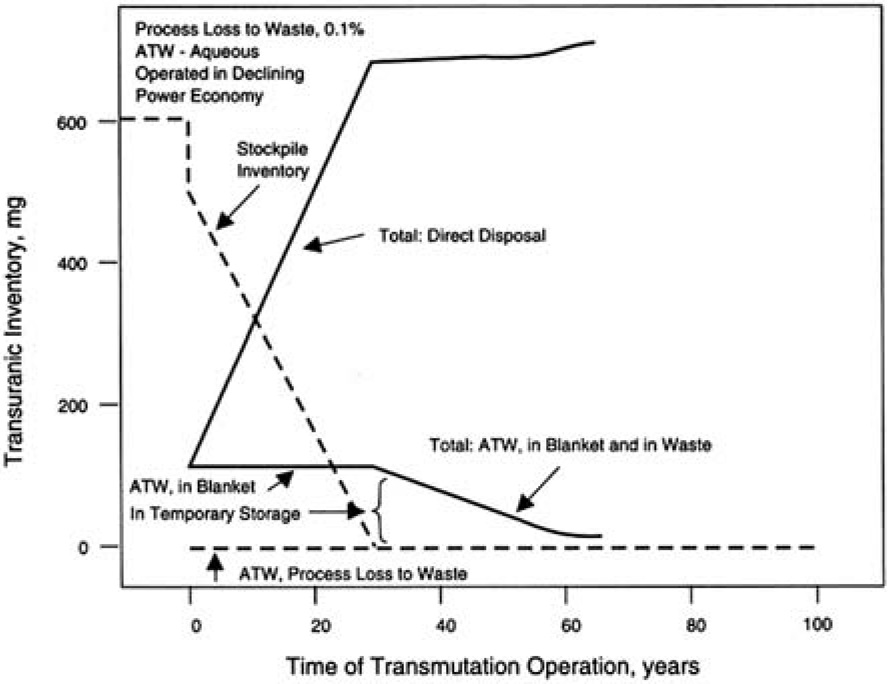

FIGURE 4-8 TRU inventory versus time for declining-power ATW.
SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

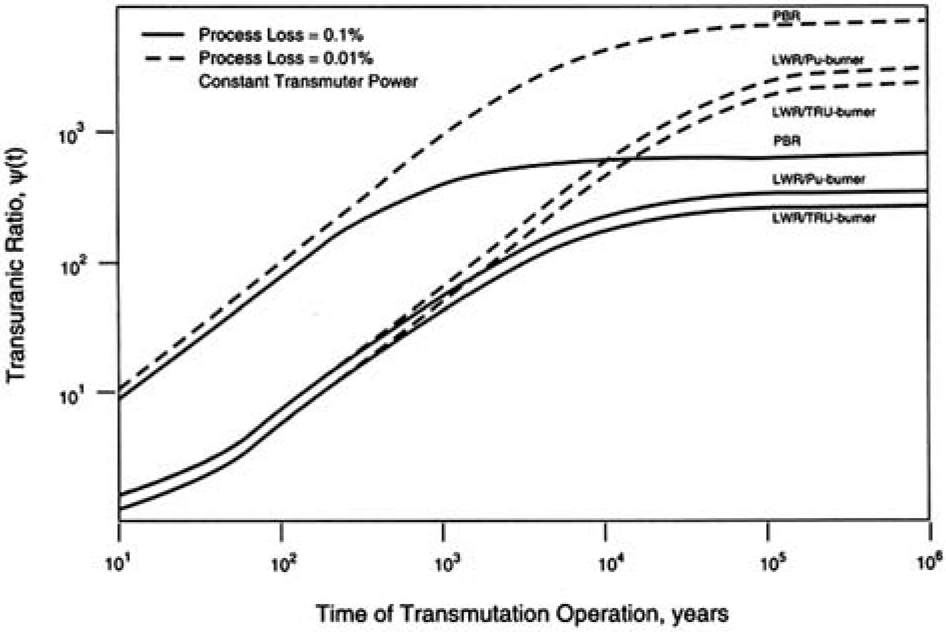

FIGURE 4-9 TRU ratio versus time for constant-power LWRs and effect of process loss.
SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

erating in self-generated recycle, with a portion of the core of each reactor fueled with recycled MOX fuel. Thus, with suitable reprocessing and MOX-fabrication capability, such transmutation could be implemented in existing LWRs. These LWRs could also be used to transmute some of the long-lived fission products, e.g., ^{99}Tc and ^{129}I, that are significant contributors to the long-term risks from geologic disposal.

MOX fuel compositions resulting from the recycle of all TRUs in a LWR transmuter approach steady state slowly, because neutron capture leads to higher mass TRUs. Achieving criticality at full steady-state is marginal and might require the use of slightly enriched uranium in formulating the recycled MOX fuel. Uranium-enriched MOX fuel could also add reactivity for transmutation of selected fission products.

Declining-Power LWR Transmuters

TRU ratios for LWR transmuters in a declining-power scenario are shown in Figure 4-5. An ultimate TRU ratio of about 100 could be achieved if the smallest practical LWR transmuter is assumed to operate at an electrical power of 600 MWe. The time to achieve a given inventory reduction is about the same for the full-recycle LWR transmuter as for the ALMR of 0.65 breeding ratio.

PBR Transmuters

TRU ratios for PBR transmuters for steady power are shown in Figure 4-9. The curve for declining power is shown in Figure 4-5. The TRU ratios and time constants are similar to those of the nonaqueous ATW, a consequence of the high fissile-specific power of the PBR.

Nonbreeding Transmuters Converted to Breeders After Stockpile Depletion

As discussed in the section on ALMR transmuters, the fuel-cycle costs of the nonbreeding ALMRs are expected to be greater than those of the breeding ALMRs, because the breeders do not depend on an external supply of TRUs for fuel make-up. This is largely a consequence of the cost of reprocessing LWR spent fuel. Consequently, constructing additional LWRs to furnish TRUs to the continuing nonbreeding ALMRs, as suggested by DOE's ALMR program, may not be an economical choice, unless the additional charges to the ALMRs are defrayed by a subsidy for waste disposal benefits. Also, in the absence of a special waste subsidy, it is questionable that the proposed strategy of a mixed system of new LWRs and nonbreeding ALMRs would be more economical than an economy based on ALMR breeders. If the future ALMRs are not basically more economical than the future LWRs, the ALMRs could not be prudently selected for electric power generation without subsidy. If the ALMRs are more economical than LWRs, the power producers should construct ALMRs and not additional LWRs. Also, limitations of uranium resources may preclude the long-term use of LWRs to fuel nonbreeding ALMRs.

Because of such economic uncertainties, a scenario involving transmutation of TRUs in the LWR spent fuel by first-generation nonbreeding ALMRs, continued by constructing later-generation breeding ALMRs, should also be considered. No additional LWRs to supply make-up TRUs would be required. The TRU ratios for this scenario involving ALMRs are shown in Figure 4-10. As pointed out earlier, the ALMR of 0.96 breeding ratio is a break-even breeder. It could ultimately achieve a TRU ratio as high as 153 because it maintains constant inventory of TRUs in the transmuter and its fuel cycle. Only a small fraction of the bred TRUs are converted to waste, whereas the reference once-through LWRs of the same power would continue to produce TRUs in its spent fuel sent to waste disposal.

Figure 4-10 also presents the TRU ratios for nonbreeding ATW's converted to breeders. Two families of curves are presented. For the upper family, the breeding ATW is assumed to incorporate rapid separation of ^{233}Pa that is then stored for decay to ^{233}U, resulting in far lower inventories of actinides than in the other ATW options. Curves involving the aqueous ATW are not shown because no breeding version of that system has been described by the designer. Again, the high specific fissile power and low inventories of the actinides result in more rapid increase in TRU ratios than for ALMRs. The higher TRU ratios of the ATWs persist for operation as long as about 1,000 years. For the assumed process loss of 0.1%, the ultimate TRU ratios (inf) of the ATWs are lower than for the ALMRs because of the more frequent processing of the reactor inventory through the coupled on-line separation system. However, if the much lower process losses expected by LANL are achieved, the long-term performance of the ATW would improve. The long-term curves for the ATW5 and ATW6 in Figure 4-7 would apply.

Constant Power Transmuters Followed by Declining Power Shutdown

The TRU ratios for constant-power transmuters are greater than those for the declining-power scenario. If constant-power operation of a transmutation system were to be suddenly terminated, it would not be necessary to send all of the TRUs in the reactor and fuel cycle to waste disposal, provided a commitment could be made to continue transmuter power in a stepwise declining mode, as illustrated in Figures 4-4, 4-5, and 4-8. The curves of Figure 4-5 indicate the extent to which inventories in the reactor and fuel-cycle at the end of constant power could be reduced by subsequent stepwise reductions in the power levels for several transmuter lifetimes.

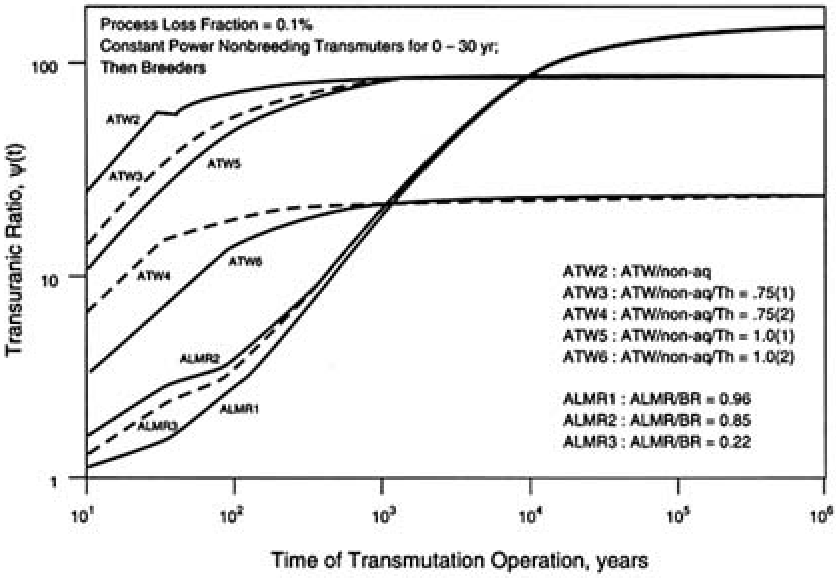

FIGURE 4-10 TRU ratio versus time for constant-power nonbreeding ATWs and ALMRs for 30 years, followed by breeders. SOURCE: Choi, J. S., and T. H. Pigford, 1994. Reduction in Transuranic Inventory by Transmutation. Report UCB-NE-4177. Berkeley: University of California.

Summary

Based on this analysis, it is clear that:

1. The inventory of TRUs[17] in a transmuter and its associated fuel cycle is appreciable and should be considered as a potential waste to be disposed.
2. The fractional loss to waste of TRUs supplied for transmutation is greater than the fractional loss to waste in each reprocessing cycle.
3. The TRU ratio (*t*) is a useful measure of the transmutation performance of a transmutation system when compared with the amount of TRU wastes that would be produced by reference LWRs of the same electrical power and energy as the transmuters.
4. For process losses of 0.1% per cycle, the maximum possible TRU ratios are a few hundred for ALMR, LWR, and nonaqueous ATW transmuters and less for the aqueous ATW. 5. The time constant[18] to achieve the ultimate constant-power transuranic ratio is many thousands of years for ALMR and LWR transmuters and a few hundred years for the ATW transmuters.
6. A declining-power scenario, involving stepwise reductions in transmuter power until an acceptably small fraction of the original TRU inventory remains, achieves more rapid inventory reduction than steady power. With declining power, the time to achieve a TRU ratio of about 100 is several centuries for the ALMR and LWR transmuters and several decades for the nonaqueous ATWs. In several decades the aqueous ATW could achieve a TRU ratio of about 50. Subsequent generations of any of these transmuters would be required. Achieving actinide reduction factors as high as 100 may be an unrealistic goal.
7. If overall TRU ratios of the order of 100 are desired to benefit waste disposal, the ratio must apply to the entire national system of nuclear power generation. Any of the transmutation scenarios considered here would require commitments to construct and operate the transmuter system and its later-generation replacements for long periods of time, of the order of centuries for declining power and many centuries to millennia for constant transmuter power. To achieve and maintain that benefit to national waste disposal, construction and operation of other nuclear power systems, such as once-through LWRs, would be limited to the number and total power specified, so that spent fuel from those reactors could produce the amount of TRUs needed as makeup for the transmuters. Institutional problems in making such commitments by government and industry are discussed in Chapter 6 of this report.

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[17] For nonaqueous ATW transmuters incorporating some thorium for internal conversion to ^{233}U, we should express the TRU ratio in terms of total actinides.

[18] The time constant is the time for the constant-power TRU ratio to reach a fraction (1 - 1/e) of the ultimate steady-state value.

REDUCTION OF KEY FISSION PRODUCT INVENTORIES

In the estimates of long-term radiological repository risk, dissolution and migration scenarios are prominent contributors. Such scenarios identify two key radionuclides as especially important on a relative dose basis, namely, the long-lived soluble fission products ^{99}Tc and ^{129}I. The predicted maximum dose of a third such fission product, ^{135}Cs, is reduced by a factor of about 360 due to sorption on the rock through which it would be transported by the groundwater. As discussed above in this chapter's section, *Transmutation Processes and Concepts* , transmutation in a nuclear reactor does not seem practical for ^{135}Cs. Its radiological risk could be decreased, if necessary, by the choice of a waste form in which cesium would have a significantly reduced solubility in groundwater.

The next most important radionuclides for dissolution-and-migration scenarios are the long-lived MAs ^{231}Pa and ^{237}Np. Many performance analyses show that their predicted maximum doses are several orders of magnitude below those of the two key fission products for dissolution and migration scenarios (see Chapter 2; Radionuclide Release Scenario). Hence, this section evaluates the transmutation proposals for their ability to achieve a reduction of several orders of magnitude in the amounts of ^{99}Tc and ^{129}I in the separated waste, i.e., to a level at which ^{231}Pa and ^{237}Np would begin to affect the long-term risk scenarios.

The transmutation of the ^{99}Tc and ^{129}I is characteristically much more effective in a thermal (or epithermal) neutron spectrum that in a fast neutron spectrum. The difference in performance (and flexibility) is due to generally higher neutron capture cross sections at lower energies. Also, the thermal (or epithermal) transmutation of ^{99}Tc and ^{129}I results in stable isotopes from neutron capture together with beta decay.

To transmute ^{99}Tc and ^{129}I, technetium and iodine would each have to be separated in the reprocessing operations. Separating ^{129}I, together with ^{127}I, from spent fuel is easily possible, although high recoveries would require some process development[19] . Separation of technetium from other platinum metals would be more difficult. About half is present as soluble residue (technetium, TcO_2) and about half as TcO_4 (Baetsle, 1993).[20]

The LWR-based transmutation concept has a high degree of flexibility for transmuting fission products. For a thermal neutron spectrum and flux level typical of a uranium-fueled PWR the transmutation rates of very dilute ^{99}Tc and ^{129}I are about 11%/yr and 3%/yr, respectively (Wachter and Croff, 1980). However, as the concentration of these nuclides is increased to achieve practical loadings in the reactor, neutronic effects reduce the transmutation rate to the point that achievable rates are expected to be about 3-4%/yr for ^{99}Tc and 1.5-2.0%/yr for ^{129}I (Kloosterman and Li 1994; Hugon 1994), with the equivalent transmutation half-lives being 20 and 40 years, respectively. These transmutation rates are marginal for a realistic transmutation system. Initial calculations of the performance of the ATW alternatives indicate a potentially attractive degree of flexibility for transmuting key fission products in its thermal spectrum.

It is possible to soften the neutron spectrum of a typical fast reactor, for example, by introducing a neutron moderator into the radial blanket. This method can obtain a spectrum that has reasonably good characteristics for transmuting the ^{99}Tc and ^{129}I at 5-10% per year with only a modest loss of efficiency for transmuting the TRUs. The price of this method, however, is a more complex reactor system. If so modified, an ALMR system would have a moderate degree of flexibility for transmuting key fission products. The previous section in this chapter also notes that it is possible to soften the neutron spectrum of a typical fast reactor, for example, by introducing neutron moderator into the fuel assembly containing the Tc or I. The CURE proposal adopts this approach and predicts transmutation rates of 5-10%/yr for ^{99}T and ^{129}I with only a modest loss of efficiency for transmuting the transuranics. More recent calculations using moderated assemblies in a fast reactor predict transmutation rates that are about the same as those given immediately above the PWR (Kloosterman and Li, 1994; Hugon, 1994). One price of this method, however, is a more complex reactor system. If so modified, an ALMR system would have a moderate degree of flexibility for transmuting key fission products.

The thermal spectrum of the PBR also should be good for burning fission products, assuming a suitable fuel form can be developed, but no work has been done to estimate the transmutation rates for ^{99}Tc and ^{129}I with the PBR. Phoenix, an accelerator-driven concept with a fast neutron spectrum, has only limited reactivity in its core because it recycles only the MAs and ^{129}I, sending the plutonium to LWRs and leaving technetium disposition an open issue. The Phoenix proposal assumes that ^{129}I would be transmuted at the same rate that it is created, but no detailed work on this aspect of the concept has been done to date. In fact, the total iodine inventory remains to be worked out.

SAFETY ISSUES FOR THE REACTOR, FUEL MATERIALS, AND SUPPORTING FUEL CYCLE

Relative to direct disposal of the spent fuel from a once-through base case, a reduction in radionuclides results in a

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[19] It is routinely evolved quantitatively from the dissolver and recovered on silver iodide.

[20] Tc is recovered by centrifugation (See D-65).

small increase in radiological hazards to the public. This increase is due to the operation of transmuters and their fuel-cycle facilities over the long transmutation period. The safety issues associated with supporting separations facilities and with transportation of the radioactive materials between sites must also be considered. In addition, the ATW target fuel contains radionuclides that emit neutrons and produce heat, presenting safety issues in fuel fabrication, handling, and qualification. This section summarizes the main issues that affect the reactor safety evaluation and related risk issues of the transmutation systems. The resulting consequences of these health and safety issues are covered in detail in Chapter 6 and Appendix I.

Decay-Heat and Target-Heat Removal

The decay heat from irradiated fuel raises important safety and licensing issues for any reactor. Spent fuel containing MAs will generate a larger amount of decay heat than conventional fuel. Passive decay-heat removal during shutdown is a crucial safety and licensing issue for all the transmutation concepts and is critical to obtaining safety approval. It is especially critical for the ATW, Phoenix, and PBR concepts that have much higher fissile-specific power, power density in the core, and levels of radioactive decay than is typical of current designs. For example, the PBR concept has almost two orders of magnitude higher power density than an LWR; an ATW operates with an order of magnitude higher thermal flux and a factor of 2.5 higher power density in the fissioning blanket than in an LWR; and the Phoenix concept has a higher fast neutron flux and power density than an ALMR. In addition to decay heat, the ATW and Phoenix concepts involve major target-heat dissipation and removal issues that affect reliable operation, as well as licensing. Analysis of the decay-heat removal and target-heat dissipation is under way for the ATW, but the issues are far from resolved. Comparatively little has been done to date on such questions for Phoenix and the PBR.

Reactivity Control

The thermal critical reactor concepts, the LWR and PBR, present issues concerning the control of reactivity and power distribution in the reactor core. This is due to the high fissile-specific power in a thermal spectrum for burning plutonium. The analysis of these issues is important for the licensability of any of the system designs. Based on work to date with self-generated recycle of plutonium in LWRs recently done by Lee and Du (1994), the extension of this analysis to cover the burning of MAs in an LWR seems possible without any difficulty. No such analysis has been done to date for the PBR.

The ALMR presents control issues peculiar to a fast critical reactor, in particular, passive reactivity reduction to a safe, stable condition for overcooling or overpower events with failure-to-scram. The use of a metallic fuel and the large thermal intertia of the pool of sodium are important in achieving safe reactor shutdown. The ALMR project finds the sodium void reactivity worth and Doppler coefficient of reactivity to be acceptable for the safety of Pu/MA burner design for values of the breeding ratio, β, near unity (Thompson, 1992). However, information available on the MA burner design is not sufficient to evaluate the reactivity characteristics as ß is reduced to 0.6. Thus, considerable experimental and calculational effort appears necessary in order to develop a database sufficient to support licensing with a core that is fueled entirely with TRUs without compromising the safety characteristics of current ALMR designs.

Reactivity Transients

The proposals for an accelerator-driven subcritical reactor claim that shutdown can be accomplished rapidly by shutting off the accelerator. For a subcritical assembly with a multiplication factor of 0.95 (ATW) or 0.98 (Phoenix), this measure presumably would be limited to the control of transients involving a reactivity swing of not more than 5% (ATW) or 2% (Phoenix). Furthermore, neither proposal provides for any additional control absorbers. The ATW and Phoenix concepts involve several types of transients, as of yet not evaluated in any detail, which raise significant safety issues that would not be eliminated by shutoff of the accelerator.

Such unevaluated transients for the ATW include: (1) reactivity swings when moving from hot operation to cold shutdown, arising from the mismatch between the expansion of the fluid fuel and the structure of the heavy-water calandria (ATW-1) or the graphite moderator (ATW-2, ATW-3, and ATW-4); (2) xenon oscillations, i.e., spatial oscillations in neutron flux and power density coupled with nonuniform xenon concentration, an effect well known in conventional power reactors; and (3) instability from ^{233}Pa - ^{233}U transients with the ATW-3 and ATW-4, which could make it difficult to shut down the reactor. Processing of the fluid fuel every 2 hours is an option under consideration that may help alleviate concern about this last issue.

Unevaluated transients for Phoenix also include sodium void reactivity, worth as well as reactivity swings when moving from hot operation to cold shutdown. Given the proposed reactivity margin of only 2% and no control rods, such transients may be even more significant for Phoenix than for the ATW. In addition, the burn-up of MAs while plutonium builds up, will bring about a major swing of reactivity for Phoenix. Estimates indicate that K_{eff} will change by 0.25 over a burn cycle, which may require a means of control beyond the accelerator. This subject has not yet been addressed.

Materials Degradation

The ATW, PBR, and Phoenix concepts present major materials degradation issues arising from operations, e.g., due to neutron-induced damage or in situ transmutation products such as helium. These conditions are beyond the experience with present-day reactors (see the discussion under the Engineering and Materials subsections in Chapter 4 and Appendix F for a broader discussion). To date, such issues have generally not been analyzed despite their impact on the reliability of the systems in continuous operation, as well as on the safety analysis required for licensing.

Fluid Fuels in the ATW

The fluid fuel in the ATW concepts poses special reliability problems that are closely related to safety. The following three examples indicate areas that could have serious implications not yet analyzed by LANL for the present designs (see Appendix F): (1) fluid boundary-layer heating with ATW-1 in the slower moving slurry near the pressure tube surface, which has been observed in previous slurry fuel systems and that may enhance the wall erosion caused by the high-density slurry; (2) pressure-tube failure, possibly enhanced by materials degradation in the high neutron flux and/or by wall erosion with ATW-1 or by corrosive action of the molten-salt carrier fluid with ATW-2, ATW-3, and ATW-4; or (3) the possibility of explosive gases released from radiolytic decomposition of the heavy-water carrier of the fuel slurry with ATW-1.

The possibility of pipe breaks is a major accident scenario for conventional reactor systems. LANL has not yet evaluated the possibility of pipe breaks in the recirculating fluid system, either inside the subcritical reactor proper or between the reactor and the separations-reprocessing equipment. For Phoenix, a pipe break in its coolant transport or heat transfer system surely represents a major accident scenario, an issue that BNL has not addressed to date.

Potential for Reduction in Mining and Milling Hazards

A possible reduction in uranium-ore mining and milling, with an accompanying decrease in occupational safety hazards (accidents and irradiation of personnel), could occur for those transmutation systems that produce enough excess plutonium from breeding to offset part of the need for fresh enriched uranium ore (e.g., an ALMR/IFR).[21] This may affect those countries where uranium ore is obtained using conventional mining and milling practices. However, such a reduction in ore requirements may have only a limited effect on the occupational health and safety of the U.S. nuclear industry workers because of the shut-down of many open pit and underground uranium mines in favor of solution mining of uranium (as well as a large increase in the fraction of imported uranium ore to meet domestic requirements). If the changes in U.S. industry practices introduced during the 1970s and 1980s are continued, a considerably smaller benefit in occupational health and safety would result from a reduction in uranium-ore mining and milling, compared with previous health and safety estimates based on the practices that prevailed in the United States prior to the 1970s (see Chapter 2 and Chapter 6 for details). Regulations governing solution mining practices must ensure that the disposal of the waste effluent from uranium mining, which contains uranium daughter radionuclides such as ^{226}Ra and ^{231}Pa, does not constitute a long-term radiological hazard.

DEVELOPMENT TIME, COST, FEASIBILITY, AND RISK THROUGH SYSTEM DEMONSTRATION

LWR Transmutation System

The most mature technology base for a S&T system is that of the LWR modified to optimize burning of TRUs and selected fission products, with aqueous reprocessing of the spent fuel and refabrication of the recovered material into fuel-rod assemblies. There are some 4,000 reactor-years of LWR operation worldwide, including experience with self-generated uranium-plutonium recycle and dedicated plutonium burners, both of which involve refabricating plutonium-uranium MOX fuel in remotely operated facilities.

However, there is neither operating nor licensing experience with multiple-recycle LWR fuels. Such target fuels contain a substantial fraction of higher-mass MAs and higher plutonium isotopes, which build up with multiple recycles in a thermal neutron spectrum. In contrast, the build-up of higher-mass TRUs to multiple-recycle fast reactor fuels is orders of magnitude lower. However, the experience with aqueous-based reprocessing of LWR spent fuel and with remotely operated fuel fabrication provides an initial basis for design and operation of the supporting fuel cycle.

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[21] An ALMR-based transmutation system does not eliminate the need for fresh uranium ore on an ongoing basis if the breeding ratio is less than unity to increase the rate of waste burn-up. Even with a breeding ratio near unity, enriched uranium may be needed for start-up, depending on the cost of start-up with reprocessed LWR spent fuel.

Using LWRs for transmutation of TRUs and fission products requires development effort (discussed in Appendix F) in three main areas: (1) refinement of the process for manufacture, quality assurance, and performance verification of target fuel containing substantial radioactivity in the refabricated fuel rods, particularly techniques to qualify target fuel that emits a substantial neutron flux; (2) refinement of control absorber effectiveness and reactivity control to accommodate the variations in reactivity as the plutonium and MA concentrations change during a cycle; (3) scale-up and pilot-scale demonstration of TRUEX (second stage) separations technology to achieve a significantly lower process decontamination factor than typical of PUREX separations alone, as well as the development and scale-up of technology for additional separations (e.g., technetium). These areas affect licensing that, in turn, affects the time scale for development and system testing. LWR-based transmutation has the shortest time and lowest cost to complete demonstration—in about 8-10 years—assuming the use of existing facilities for in-reactor fuel testing and demonstration, and exclusive of any large-scale separations demonstration plant, if required. The system is also expected to have the lowest development risk and the lowest cost to complete demonstration (estimated at perhaps $50-$100 million per year; see Appendix F [E. Evans, private communication, 1993]) in view of the considerable background of worldwide experience in the use of plutonium bearing fuels in LWRs.

ALMR/IFR Transmutation System

The ALMR component of the ALMR/IFR system has been under development for decades as a breeder of fissile nuclear material. Several such reactors have been operated in the United States and aborad. There is substantial design and operating experience with full-scale fast reactors using metallic and MOX fuels containing uranium and plutonium. Since actinide transmutation in an ALMR would operate with a different fuel and operational regime than usual, further development and proof testing would be required in two main areas: (1) reactor safety and control, especially if the ALMR is to be operated at a breeder ratio appreciably less than the normal value of 1.05 in order to increase the rate of TRU inventory reduction; and (2) fabrication, quality assurance, and performance verification of fuel containing TRUs (see Appendix F). Qualifying the target fuel raises issues similar to those discussed above for LWR transmutation, albeit with much lower levels of radioactivity in the ALMR target fuel. The details would depend on whether the ALMR uses conventional fuel pins fabricated with oxide or metal fuel elements, or fuel fabricated as part of integral pyrometallurgical reprocessing (see below). Both areas (1) and (2) affect licensing that, in turn, affects the time scale for development and system testing.

DOE proposes an ALMR/IRF transmutation system for TRUs. This comprises ALMRs collocated with integrated pyrometallurgical capability for in situ processing of ALMR spent fuel and fabrication of recycle fuel. The IFR replaces a centralized aqueous-based reprocessing capability previously studied by GE for the ALMR. ANL has demonstrated the pyroprocessing of mock ALMR metallic fuel at laboratory scale but has not yet demonstrated the process at pilot-plant scale. ANL has also not yet demonstrated the related process with LWR oxide-spent fuel, even at laboratory scale. Because the operation with pyrometallurgical reprocessing is substantially different from that of any licensed system, a full-scale system demonstration would no doubt be required. The ALMR/IFR alternative is expected to have an intermediate development risk, cost, and an intermediate time between LWR's and ATW's to complete demonstration—about 15 to 20 years, assuming the use of existing facilities for in-reactor fuel testing and demonstration, exclusive of the cost of any full-scale demonstration facilities.

Indeed, because a low threshold cost for LWR spent-fuel reprocessing is required for an economically viable ALMR/IFR TRU burner, its development risk is higher than that of an ALMR for power production. The development risk would be further increased if a large centralized pyroprocessing facility were to be adopted for LWR spent fuel, as proposed by DOE/NE, rather than a facility based on aqueous reprocessing technologies.

Accelerator Transmutation of Waste (ATW)

The ATW concepts use an accelerator to provide neutrons from a spallation target that are moderated to thermal energies and multiplied in a subcritical reactor. The very high neutron flux levels and the energy density in the core, and the associated decay heat and nuclide destruction, are well beyond the experience with conventional thermal reactors. Thus, the ATW concepts have attractive projected transmutation performance but present major engineering and materials development challenges, especially in the beam target, subcritical reactor, and recirculating fluid-fuel subsystems.

Because of the progress that has been made in the technology for high-energy accelerators with high beam current, it is likely that the accelerator current-energy requirements for the ATW transmutation concepts could be met with perhaps a decade of additional development, testing, and demonstration with a large-scale prototype. The major challenge for the accelerator is the requirement for higher performance and reliability in continuous operation than heretofore achieved (see the detailed discussion in Appendix F).

However, only limited reactivity swing is available for control by shutting down the accelerator, about 5% for the present ATW concepts. This report notes several possible reactivity transients not yet studied by LANL that may well require provision for additional reactivity control. Yet, none of the ATW concepts include control absorbers. Further study of the reactivity control issues by LANL may lead to system design changes with additional development and testing requirements.

Indeed, for the ATW the possible need for control rods and the related reactivity control system raises questions about the benefit of using an accelerator and subcritical neutron-multiplying-moderating assembly, compared to a high-performance, just-critical thermal reactor with conventional reactivity control. Moreover, there is an economic penalty for an accelerator, which is substantially greater for the aqueous ATW-1 than for the other three ATW concepts. Problems due to materials degradation in the high neutron fluence and to the fluid fuels (summarized below) would be essentially the same, with or without the accelerator. Thus, LANL may find that eliminating the accelerator and beam target and going to just-critical reactor concepts could result in simpler core designs and more economical systems.

The ATW concepts present materials degradation issues, largely unanalyzed at present, that are likely to require extensive materials development and testing (see Appendix F). Two major types of such degradation are (1) radiation damage in the target and blanket assembly structure, especially from neutrons; and (2) alteration of the mechanical properties and dimensional stability of structural materials caused by transmutants, in particular, by helium from alpha particle release. These issues present significant problems in conventional reactors and would be more severe under the ATW operating conditions.

The subcritical reactor assembly and proton beam target for any of the four ATW concepts pose major engineering and materials development challenges (see Appendix F). The beam targets are to be cooled with molten lead (ATW-1) or molten lithium (ATW-2, -3, and -4) that pose substantial heat removal and materials issues under the anticipated conditions. In addition, the slurry fuel for ATW-1 and the molten salt fuel for the other ATW concepts raise significant materials issues not yet studied by LANL, as described in an earlier section of this chapter. Both the slurry fuel and molten salt concepts were the subject of development efforts two decades ago, and major materials problems associated with various fluid-materials interactions, including corrosion were found. In addition, the system performance depends critically on the high recovery separations-reprocessing system, many aspects of which have not yet been demonstrated beyond the laboratory scale.

Extensive reactor, target-fuel, and fuel-cycle development and performance verification would be required for any of the ATW proposals before the chances of success could be determined. Given success in the development stage, both pilot-scale and full-scale test and demonstration would be required. ATW concepts (aqueous and nonaqueous) require the longest time to complete full-scale demonstration—an orderly program may take more than two decades through complete demonstration at a cost of perhaps $1 billion or more. They also carry a high development risk (see Appendix F).

Other (Secondary) Transmutation Concepts

The PBR and the Phoenix transmutation systems are still at the conceptual study and design stage. Like the ATW, these systems entail very high neutron flux levels and associated heat generation and nuclide destruction rates that are beyond the operational experience with conventional present-day reactors. Because only a scant amount of information to evaluate the two concepts in available, only a limited evaluation is presented here.

PBR Transmutation System

The PBR is a critical reactor concept with a very high thermal flux that could achieve rapid burn-up of TRUs and fission products, assuming a suitable fuel form could be developed. However, PBR is based on a large extrapolation of the technology being developed for a space reactor. PBR has a much higher fissile-specific power, power density in the core, and levels of radioactive decay than are typical of current thermal reactor designs. In fact, the PBR conceptual design has almost two orders of magnitude higher power density than a conventional LWR. The advanced PBR operating conditions raise a host of major materials and engineering issues, for the reactor itself and for target fuels (see Appendix F).

The following list of PBR development challenges are illustrative: (1) a highly reliable helium coolant system for the packed particle bed, to cope with the extraordinary heat generation rate and core power density; (2) requirements for the Be_2C moderator blocks and the structural materials to withstand fluences of the order of 10^{23} neutrons/cm^2; (3) target fuel comprising TRUs and fission products embedded in pyrographite particles, reminiscent of HTGR fuel particles but much smaller in diameter, and able to withstand a limiting temperature of 2,200°C while retaining fission products (considerably higher than the 1,600°C limiting temperature for HTGR fuel); and (4) a modified PUREX-TRUEX process plus a lanthanide separations stage, with an electrodissolver at the front end to remove the carbon; a fluoride volatility process is being considered as an alternative, but that process has been plagued with problems in past applications.

Extensive reactor, target-fuel, and fuel-cycle development and performance verification would be needed before even the chances of success could be assessed. Because of the substantial amount of technology development, testing, and pilot-and full-scale demonstration required to meet its ambitious goals, the PBR concept has high development risk. An orderly program may require about two decades through full-scale demonstration at a comparatively high cost, totaling perhaps $1 billion or more (see Appendix F).

Phoenix Transmutation System

The Phoenix fast neutron transmutation concept has accelerator requirements much like those discussed above for the ATW. The accelerator current-energy requirements look achievable with further development, provided high performance and reliability can be attained in continuous operation. However, only limited reactivity swing is available for control of the subcritical reactor by shutting down the accelerator, about 2% in the present Phoenix concept, with no ancillary control absorbers. This report notes several possible reactivity transients not yet studied that may well require provision for additional reactivity control. This could lead to system design changes with additional development and testing requirements.

Moreover, the Phoenix subcritical reactor would operate at conditions beyond the experience with present-day fast reactors. Thus, Phoenix presents significant materials degradation issues, unanalyzed at present, from radiation damage and from alteration of the mechanical properties and dimensional stability of structural materials caused by transmutants. These issues would require extensive materials development and testing (see Appendix F).

Phoenix proposes to use the MOX fuel as the beam target, as distinct from the ATW concepts that use a separate target. The fuel would be similar to that tested in FFTF. However, there is neither analysis nor experimental results as yet to support the extension of the previous work with MOX fuel to the requirements for long fuel life while burning MAs in the Phoenix spectrum and flux levels. This also raises similar fuel qualification and performance verification issues as discussed above for LWR transmuter fuel, albeit with much lower levels of neutron emission in the Phoenix target fuel. In addition, cooling the Phoenix fuel with molten sodium raises significant materials issues under the anticipated conditions. Finally, the Phoenix proposal assumes a developed PUREX-TRUEX separations process similar to that discussed for the LWR transmuter but with a very high process decontamination factor, i.e., 10^5, stated as a goal by BNL.

Like the ATW, extensive reactor, target-fuel, and fuel-cycle development and performance verification would be required before even the chances of success for Phoenix could be determined. Given success in the development stage, both pilot-and full-scale demonstrations would be required. Although Phoenix draws on oxide fuel experience, its development risks are high, and the time and cost requirements through complete full-scale demonstration may be comparable to developing one of the ATW concepts.

TIME SCALES AND COSTS FOR MODEL SYSTEM DEPLOYMENTS

Basis for Transmutation System Deployments

This section summarizes deployments of selected S&T systems in terms of material quantities, costs, and time scales. For illustration, we discuss the scenarios of declining nuclear power and of constant nuclear power generation (see section on Reduction of Transuranic Inventories).

Material Flows. Table 4-2 presents inventories of TRUs in the reactor and external fuel cycle for nuclear power plants operating at 1,395 MWe. It presents ratios and other data from which annual material flow quantities can be calculated. The inventories and flow quantities can be assumed to be proportional to rate of electrical generation. For example, a 1,395-MWe LWR operating as a TRU burner would have a transuranic inventory of 21,600 Mg and an annual discharge rate of 38 Mg HM/year.

Reprocessing Costs. Appendix J analyzes cost information available on several foreign facilities that use a PUREX-like separations technology to reprocess LWR spent fuel, for a once-through fuel cycle. Results are summarized in Chapter 6. The facilities analyzed include the French UP3 plant, the British THORP plant, and the new Japanese facility under construction at Rokkashomura. These data are used to estimate capital and operating costs of a technically comparable facility of 900 Mg/yr nominal annual operating capacity, if constructed in the United States. The projected capital cost in 1992 dollars is shown in Table 4-5.

The capital cost includes facilities for temporary storage of spent fuel and a facility to convert the HLW to a suitable form for geologic disposal. The operating cost is projected at $380 million/yr. The resulting 30-year levelized unit reprocessing cost for U.S. construction and financing is shown in Table 4-5.

TABLE 4-5 Projected capital reporcessing costs in 1992 dollars

Financing	Capital cost, $ billion	Unit cost, $/kg HM
Government	6.2	800
Utility	6.7	1,300
Private	7.3	2,100

The above cost estimates are for a facility processing LWR discharge fuel from a once-through fuel cycle, with process losses characteristic of current PUREX technology. Additional technology, such as the TRUEX process, would be required to reduce process losses to the level of about 0.1%, as sought by DOE contractors for transmutation. For illustration, we adopt a 10% increase in the above costs for a high-recovery PUREX-TRUEX facility. Costs for reprocessing MOX fuel are certain to be much higher, but we have no estimates of the magnitude. Consequently, we adopt the above costs increased by 10%.

The reference reprocessing facility with a throughput of 900 Mg HM/year could reprocess spent fuel discharged from 24 1395-MWe LWRs.

The total cost to reprocess the 62,000 Mg HM of spent LWR fuel now destined for Yucca Mountain would be from $50 billion to $130 billion for a U.S. reprocessing facility, depending on the method of financing. In addition to the reprocessing of the spent fuel, reprocessing of the fuel discharged from the transmuting reactor would be required. The amount of such fuel being reprocessed is about a factor of 10 less than the LWR spent fuel. Further, all the LWR fuel would have to be reprocessed by the end of the first 30-year cycle. The processing of the transmuting reactor fuel is required over a much longer time scale. If we discount the cost of this reprocessing of transmuting reactor fuel, its cost becomes small compared to the cost of reprocessing the spent LWR fuel.

For the ATW, LANL plans fluid fuel systems close-coupled to on-line reprocessing plants. The proposed technology for the ATW-1 is a slurry fuel of small TRU particles suspended in a heavy-water carrier, circulating in small pressurized tubes inside the reactor. For the ATW-2, LANL proposes a molten salt carrier with dissolved TRU radionuclides, again circulating in tubes. However, the reprocessing system is not yet defined for either version—indeed, major technical development is required—so no reliable basis exists to estimate ATW reprocessing costs.

Reactor Costs. The approach to reactor costs is adapted from the National Research Council report, *Nuclear Power* (1992). This report evaluates capital costs for current LWRs and describes the Electric Power Research Institute (EPRI) goals for capital cost and operating costs for advanced LWRs of 1.2- to 1.3- GWe capacity and passively safe LWRs of 600 MWe capacity (see National Research Council, 1992: Table 3-2, p. 94). The goal for the advanced 1.2- GWe LWR is an overnight capital cost of $1,300 per rated kWe, with goals for fixed and incremental operating cost of $61.1/kWe-yr and 0.11 cents/kWh, respectively. For the 600-MWe passively safe LWR, the corresponding goals are $1,475 per rated kWe, $72.7/kWe-yr and 0.15 cents/kWh, respectively.

In addition, the report lists cost goals for ALMR-type reactors based on an estimate by EPRI (see National Research Council, 1992: Table 3-4, p. 139). The overnight capital cost goal is $1,725 per rated kWe with fixed and incremental operating cost goals of $75.5/kWe-yr and 0.15 cents/kWh, respectively. For the three types of reactors, the report quotes an EPRI uncertainty estimate of -30 to +80 percent, presumably with greater uncertainty for the ALMR than for the LWRs.[22] There is no reliable basis as yet for estimating the costs of any of the ATW transmutors.

Strictly speaking, these costs for reactors and the costs of reprocessing need to be put on the same basis. In practice, this would not only be difficult but pointless in view of the large uncertainty in the costs of reprocessing (see Chapter 6). Thus, the distinction is ignored for the costs of the model deployments. For an advanced LWR this evaluation uses a 30-year cost of $3.36 billion, comprising $1.30 billion in capital cost, $1.83 billion in fixed operating cost, and $0.23 billion in incremental operating cost (assuming 80% capacity factor). Similarly, for a 1-GWe ALMR, this section uses a 30-year cost of $4.31 billion, comprising $1.725 billion in capital cost, $2.265 billion in fixed operating cost, and $0.316 billion in incremental operating cost. Utility financing would entail additional cost.

Deployments for Declining Nuclear Power

Reprocessing Requirements and Deployment Rate

The deployments for declining nuclear power are designed to transmute the 612 Mg of TRUs in the 62,000-Mg HM stockpile of spent LWR fuel. This section compares deployments for the LWR, ALMR ($\beta = 0.65$), and the ATW systems. For a rapid reduction in total transuranic inventory, the ALMR with $\beta = 0.98$ is not a suitable candidate. The first-generation transmutors are to operate at constant power for 30 years. There are to be a sufficient number so that all of the 612 Mg of TRU in the 62,000 Mg of LWR spent fuel will be used for start-up inventory and for TRU make-up during the first generation. The reactor and fuel-

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[22] The National Research Council Energy Engineering Board (1992: Table 2-4) lists the overnight construction costs in 1988 dollars for selected U.S. nuclear power plants, by year of commercial operation. For 10 LWRs that came into operation during 1987-1988 at an average capacity of 1,070 MWe, the cost per kWe varied from a high of $4,596 to a low of $1,342, with an average of $3,133. In the previous period of 1985-1986, 15 plants came into operation with similar high and low values and an average of $2,620 per kWe. The wide variance in cost in a given time period and the high average cost (in constant dollars) compared to the 1970s are presumably due to the time and cost to meet changing regulatory requirements following the accident at Three-Mile Island, together with the high interest rates of the early and mid-1980 period. This evaluation assumes that such factors do not continue for future reactors.

cycle inventory remaining at the end of the first generation is to be used to start and fuel a smaller number of second-generation transmutors. Similar reductions in number of transmutors are made in each subsequent generation until the desired net reduction in transuranic inventory is achieved.

Using the data in Table 4-2, an LWR transmutor generating 1.395 GWe requires a start-up inventory of 21.6 Mg of TRUs and a TRU makeup rate of 0.350 Mg/yr. A total of 21.6 such first-generation transmutors would use all of the 612-Mg TRU from the stockpile during a 30-year operating life. To supply the yearly makeup of TRU, 1,097 Mg of LWR spent fuel from the stockpile must be reprocessed yearly. Reprocessing MOX spent fuel discharged from the transmutors would require an additional reprocessing capacity of 46.5 Mg HM/yr. Technology for reprocessing multiply recycled MOX fuel has not been implemented commercially. However, supplying the start-up inventory places a much greater demand on reprocessing capacity. A total of 29,100 Mg of spent fuel must be reprocessed. This would take 32 years for a single 900-Mg/yr reprocessing facility. Seven such reprocessing plants could reduce the reprocessing time to less than five years. These results are shown in Table 4-6.

TABLE 4-6 Calculated Number of Transmutors and Reprocessing Rates, to utilize Total TRU in 62,000 Mg of LWR Spent Fuel[a]

TRANSMUTOR	GENERATION AND MODE OF OPERATION	NUMBER OF TRANSMUTORS[b]	NUMBER OF LWRs[c]	TOTAL SYSTEM ELECTRICAL POWER, GWe	FRACTION OF INITIAL TRU INVENTORY REMAINING[d]	LWR SPENT FUEL REPROCESSING RATE, Mg/yr[e]		TRANSMUTOR RECYCLE REPROCESSING RATE, Mg TRU/yr[f]
						Start-up	Make-up	
LWR	First Generation	13.3		18.6	0.47	5,820	1,097	46.5
	Declining Power, Second Generation	6.2		8.7	0.22			21.9
	Continuous Power, Second Generation	7.6	5.7	18.6	0.27		208	26.5
ALMR (β=0.65)	First Generation	21.8		30.4	0.50	6,180	1,040	69.9
	Declining Power, Second Generation	10.8		15.1	0.25			34.7
	Continuous Power, Second Generation	13.9	7.9	30.4	0.32		287	44.6
ATW-1 AQUEOUS	First Generation	7.32		10.2	0.042	2,300	230	780
	Declining Power, Second Generation	1.7		2.4	0.0018			107
	Continuous Power, Second Generation	2.0	5.3	10.2	0.052		192	217
ATW-2 NON-AQUEOUS	First Generation	16.3		22.7	0.016	1.98	2,030	117
	Declining Power, Second Generation	0.26		0.36	0.0002			1.90
	Continuous Power, Second Generation	3.7	12.6	22.7	0.0036		454	27.7

[a] Contains 612 Mg of transuranics (TRU).

[b] Each transmutor operates for 30 years at 1.395 GWe (see Table 4-2).

[c] The number of LWRs required to supply TRU make-up at constant total electric power. Each LWR operates at 1.395 GWe.

[d] Ratio of the TRU inventory in the transmutors and fuel cycles, at the end of the generation, to the original 612 Mg of TRU.

[e] The rate is calculated for total "heavy metal" (HM). A 5-year reprocessing campaign is assumed for the start-up inventory.

[f] Rate at which TRU in spent fuel is discharged from transmutors, Mg TRU/yr.

By the end of the first generation, 53% of the initial transuranics will have been consumed. The remaining 287

Mg of transuranics in reactor and fuel-cycle inventory could be used to start and fuel 6.2 second-generation transmutors. No further LWR-fuel reprocessing would be required in this declining power scenario. In this manner, each subsequent generation would consume 53% of the TRUs remaining after the previous generation. Finally, after a little more than seven generations and after more than 180 years, about 1% of the original transuranic inventory would remain. Subsequent transmutation would require only a portion of the one reference transmuting reactor or smaller reactors. The unit cost of reprocessing will be increasingly expensive during later generations because of the smaller throughput.

An ALMR (breeding ratio = 0.65) generating 1.395 GWe requires a start-up inventory of 14 Mg of transuranics and a yearly make-up of 0.47 Mg/yr. Twenty-two such transmutors could utilize the entire stockpile inventory of 612 Mg TRU during their 30-year operating life. About half of the original stockpile inventory of TRU would remain. To supply the yearly makeup would require reprocessing 1040 Mg/yr of LWR spent fuel. To supply the start-up inventory, 30,900 Mg of LWR spent fuel would be reprocessed, requiring 3.4 plants, each with capacity of 900 Mg/yr of LWR spent fuel. Reprocessing ALMR spent fuel would require a special facility capable of reprocessing fast-reactor spent fuel, with an annual total capacity of about 200 Mg of ALMR spent fuel. This is less than the required capacity for a LWR transmutor because of the higher enrichment of fast-reactor spent fuel. ANL proposes to construct facilities for reprocessing and recycling ALMR spent fuel that are located at each reactor site. Each such facility would have an annual throughput of about 10 to 20 Mg of ALMR spent fuel, depending on the number of transmutors at each site. Second-generation transmutors would consist of 10.8 similar ALMRs.

The nonaqueous ATW generating 1.395 GWe has much lower transuranic inventory than the LWR and ALMR of the same generating capacity (cf. Table 4-2). The transuranic make-up rate of 1.23 Mg/yr for each transmutor, for the nonaqueous ATW without thorium, is much higher than for the LWR and ALMR, because this ATW has no internal breeding of fissile material. A much higher fuel-cycle cost can be expected. During a 30-year operating life a total of 16.3 such ATW's could utilize all of the 612 Mg of TRU in the LWR spent fuel stockpile. A total of 2030 Mg of LWR spent fuel would be reprocessed yearly to supply the make-up TRU. The reprocessing capacity to supply the start-up TRU would be more modest. It could be obtained by operating the 2,030 Mg/yr facility for about 6 months. Each of the ATW's would have its own integrated on-line reprocessing facility, capable of reprocessing 7.2 Mg/yr of transuranics, with a total capacity of 118 Mg TRU/yr for all transmutors. At the end of the first generation of transmutors only 1.6% of the original TRU inventory will remain. For the second generation a single ATW scaled down to 360 GWe would suffice.

The performance of the aqueous ATW is more modest. The large start-up inventory of 15.5 Mg of TRU for a 1.395-GWe transmutor is a consequence of the very low thermal efficiency. The transuranic make-up rate is 2.27 Mg TRU/yr for each transmutor. During a 30-year operating life a total of 7.3 such transmutors could utilize all of the 612 Mg of TRU in the LWR spent-fuel stockpile. The TRU make-up would require reprocessing 230 Mg/yr of LWR spent fuel. A total of 11,500 Mg of LWR spent fuel would be reprocessed to supply the start-up inventory. To process this in 5 years would require 2.6 reprocessing facilities, each of capacity 900 Mg/yr of LWR spent fuel. At the end of 30 years, 23 percent of the initial TRU inventory would remain. This could start and fuel 1.7 similar transmutors for the second generation.

Deployments for Steady Nuclear Power Generation

Using transmutors in a system of constant nuclear power generation has been described earlier (cf. "Reduction in Transuranic Inventory"). First, we summarize the material quantities based on a system of ALMR transmutors, with breeding ratio of 0.65, and once-through LWR reactors to supply the TRU make-up for the transmutors after the stockpile of LWR discharge fuel is consumed. This system could begin with first-generation transmutors in the same way as the first-generation transmutors for the declining power scenario. The parameters for that first generation are given above. In the second generation, rather than decreasing the total nuclear power as in the declining power scenario, additional ALMRs and LWRs will be installed so that the total electrical power is unchanged from the first generation. The ratio of additional ALMRs to additional LWRs will be calculated so that no additional TRU will be needed from an outside source.

Using the data in Table 4-2, we calculate that in the second generation there will be 13.9 ALMRs and 7.9 LWRs, each generating 1.395 GWe. The reprocessing requirements for the first generation would be the same as calculated for ALMRs in declining power. During the second generation, newly created LWR discharge fuel must be reprocessed at a rate of 287 Mg HM/yr, a rate far below current commercial scale of 900 Mg HM/yr. At the end of the second generation the remaining TRU inventory will be 194 Mg, which is 36% of the inventory remaining at the end of the first generation. Thus, continuing at constant power reduces the burnup fraction per generation well below the 50% burnup per generation that can be achieved in declining power.

In each subsequent generation the ratio of LWR power to ALMR power increases. It reaches an asymptotic value of 1.3. At this ratio, LWR spent fuel would be reprocessed at a rate of 450 Mg/yr.

The reprocessing requirements for the first-generation ATWs have been described in the previous section. During the second generation newly created LWR discharge fuel must be reprocessed at a rate of 192 Mg HM/yr. The asymptotic ratio of LWR power to aqueous ATW power is 6.3, corresponding to a reprocessing rate of 230 Mg HM/yr of LWR spent fuel.

Similarly, for the second-generation nonaqueous ATW transmutors, 454 Mg HM/yr of spent fuel from 17.4 GWe of LWR's must be reprocessed to fuel the transmutors. The asymptotic ratio of LWR power to nonaqueous ATW power is 3.42, corresponding to a reprocessing rate of 192 Mg HM/yr of LWR spent fuel. The aqueous ATW transmutor can transmute transuranics at almost twice the rate of that of the nonaqueous ATW, both at the same electrical power, because of the much lower thermal efficiency of the aqueous ATW.

Table 4-6 lists characteristics for second-generation systems of various transmutors and once-through LWRS, with total system power the same as in the first generation.

COMPARATIVE THERMAL AND ELECTRICAL EFFICIENCIES

All of the transmutation proposals except the particle bed reactor presently envision producing and selling electrical energy as a means of paying for the cost of the transmutation system. That cost includes the capital cost of the transmutor facility, the cost of reprocessing LWR spent fuel to be transmuted, and the cost of reprocessing and refabricating the transmutor target fuel itself. Setting aside Phoenix, which is a hybrid concept using LWRs to burn the plutonium, it is useful to compare the net thermal-to-electrical efficiencies of the various transmutation concepts assuming that power is delivered to the electrical grid to offset part of the cost of the system. In the present LANL design for the aqueous ATW-1 concept, in which the accelerator consumes a significant fraction of the electrical energy produced, the net thermal-to-electrical efficiency is only 19.3%. This compares to 33-36% for the nonaqueous ATW concepts and to a similar range of efficiencies for the LWR and the ALMR/IFR concepts.

If the ATW-1 concept were to operate as a just-critical reactor without an accelerator, the net thermal efficiency would be about 30%, 50% higher than for the accelerator-driven ATW-1 concept. From discussions with ATW designers, it appears that the aqueous ATW-1 as proposed could be modified by eliminating the accelerator and increasing the effective neutron multiplication factor from 0.95 to 1.0 to achieve criticality (see Appendix F). One option is to add another blanket module. Another is to increase the density of TRUs in the slurry. Yet, to achieve the 19.3% efficiency of the present ATW-1 design, LANL has already increased the fluid density and pressure to the maximum possible extent; further increase of efficiency may not be practically achievable.

For a rough estimate of the savings in total cost, Appendix F considers a campaign to process and transmute all the TRUs in the 62,000 MgHM of LWR spent fuel accumulated by the year 2011, otherwise destined for a geologic repository. Assuming that a just-critical reactor for the ATW-1 would cost roughly the same as the subcritical reactor plus target assembly in the present concept, and that electrical energy can be sold to the grid for about $0.02 per kWe, the cost differential (savings) in changing from an accelerator-driven to just-critical ATW-1 would be about $2.8 billion for the accumulated LWR spent fuel. Additional savings would accrue from not having the accelerator. Clearly, the ATW-1 pays heavily for using an accelerator, if the sale of electrical energy is expected to contribute significantly to funding the construction and operation. To realize the potential advantages of using aqueous reprocessing technology, which is much more highly developed than the technology used in the nonaqueous ATW, the features of a just-critical aqueous ATW-1 without accelerator should be carefully examined by LANL.

PRINCIPAL FINDINGS AND CONCLUSIONS

1. *The S&T of TRUs and certain long-lived fission products in spent reactor fuel is technically feasible and could, in principle, provide benefits to radioactive waste disposal in a geologic repository. However, to begin to have a significant benefit for waste disposal, an entire S&T system consisting of many facilities would have to operate in a highly integrated manner from several decades to hundreds of years. The deployment of an S&T system that is extensive enough to have a significant effect on the disposition of the accumulated LWR spent fuel would require many tens to hundreds of billions of dollars and take several decades to implement.*

1a. An S&T system using reactors having a thermal neutron spectrum, such as the LWR or ATW, could transmute the ^{99}Tc and ^{129}I in spent fuel. This in turn could reduce the calculated long-term radiological hazard of a repository in waste dissolution-and-migration scenarios. The ALMR, with its fast neutron spectrum, would not be suitable for such transmutation. As an alternative, the technetium and iodine could be separated and packaged in low-solubility waste forms for geologic disposal. Separation and transmutation of ^{99}Tc and ^{129}I would not be feasible unless a full-scale reprocessing program at a commercial level were in place.

1b. Transmutation of the TRUs could be accomplished in an LWR, an ALMR, or an ATW, assuming the latter system concept proves to be technically feasible (see 3c below). Such transmutation is one of several means (some of which do not involve S&T, of increasing the waste disposal capacity of a repository, deferring the need for a second repository (see Chapter 6). Reduction of the TRUs could also reduce the calculated radiological hazard of human intrusion scenarios in which nuclear waste is brought directly to the surface. However, transmutation of the TRUs would have little effect on the calculated long-term hazards in waste dissolution-and-migration scenarios, except possibly for ^{237}Np in some scenarios.

1c. Alternatively, the spent LWR fuel could be reprocessed and the plutonium recycled to LWRs or ALMRs (see 2h below). Compared to the once-through LWR fuel cycle, the main benefit to waste management would be the use of improved, low-loss waste forms for the separated MAs and other key constituents, such as ^{14}C, ^{129}I, and ^{99}Tc. This assumes that the improved waste forms could be developed and shown to be of greater integrity to spent fuel under the regulatory requirements for geologic disposal. In principle, transmuting the plutonium would reduce somewhat the radiological risks of the human intrusion scenarios in which nuclear waste is brought directly to the surface. The untransmuted 433-year ^{241}Am, however, would dominate the TRU activity for postclosure times from a few hundred years to several thousand years. Plutonium transmutation, therefore, may have limited impact on repository risks.

1d. The initial commitment to reprocess the 63,000 MgHM stockpile of LWR spent fuel accumulated by 2011—which would require a reprocessing throughput of 2,100 MgHM spent-fuel per year over 30 years—would require an early decision to organize and carry out the development and demonstration of a prototype S&T system. Such a system would comprise many interdependent components, including waste transmutation reactors, spent-fuel reprocessing plants, recycle fuel fabrication plants, facilities to package the residual waste for ultimate disposal, and mechanisms for transportation between the facilities. Successful demonstration would be followed by the construction and operation (including maintenance and retirement) of tens of reactors and their associated fuel-cycle facilities, entailing an ongoing commitment for several generations (about 30 years each) of facilities.

1e. Merely developing, building, and operating the individual components of the system would give little or no benefit. To have a real effect, an entire system of many facilities would be needed in which all the components operate with high reliability in a synchronized fashion for many decades or centuries (see 2 below). System viability could be maintained only if the right facilities were built and put into operation at the right times. The magnitude of the concerted effort and the institutional complexity (involving long-term linkages among many private and governmental organizations) are comparable to large military initiatives that endure for much shorter periods than would be required for an S&T system.

1f. An estimate can be made of the time scale and the cost to deploy an S&T system based on LWRs or ALMRs, assuming that development can be completed (see 3 below) and that favorable institutional arrangements can be achieved. The licensing, construction, and initial operation of an S&T system of sufficient scale to *begin* to affect spent-fuel emplacement in a geologic repository would require one to two decades after a system feasibility demonstration and an expenditure of $20 to $40 billion beyond the costs of development and demonstration.

1g. Additional time and a much larger investment of funds would be necessary for an S&T system of sufficient scale to reduce repository hazards significantly or to affect the need for a second repository. For example, to reprocess the 62,000 MgHM of LWR spent fuel and transmute its 612 Mg of TRUs under a declining nuclear power (phase-out) scenario, an ALMR/IFR system operating with a 0.65 breeding ratio or an LWR system with aqueous reprocessing would cost some $500 billion and require approximately 150 years to accomplish the transmutation (see 2d and 2e below for details). Such a system, whether federally or privately owned, would not be economical in the United States at the current costs of uranium ore without a large subsidy (between $30 to $100 billion). For the same declining nuclear power scenario, an ATW deployment would require about 50 years for the transmutation (see 2f below for details). Information is insufficient to reliably estimate the cost of ATW deployments.

2. *The proposed S&T systems require decades to centuries to achieve a significant net reduction in the total TRU inventory relative to that of a once-through LWR fuel cycle of the same electrical production capacity. Comparing S&T systems using LWRs, ALMRs, and ATWs of the same electrical production capacity, the ATW projects the highest TRU consumption rates. TRU consumption rates for an ALMR increase significantly as its breeding ratio is lowered to 0.65 from the conventional value near unity. The LWR would have a net TRU consumption rate similar to an ALMR of 0.65 breeding ratio. As an alternative to S&T of all TRUs, reprocessing the LWR spent fuel and recycling only the plutonium to LWRs or ALMRs —using improved low-loss waste forms for the other HLW constituents—*

would be simpler and somewhat less expensive to implement (see 1c above).

2a. With an S&T system, each succeeding generation would transmute the TRUs left over from the previous generation plus make-up. In a continuing nuclear power economy with steady electrical production, the total TRU inventory (i.e., in the transmutors, fuel-cycle facilities, and waste) would gradually increase due to process losses. In contrast, with declining nuclear power the total inventory of TRUs in the S&T system could be reduced if each succeeding generation operated with less total transmutor power (i.e., fewer transmutors and/or lower power per transmutor). For example, each generation over its lifetime (around 30 years) could transmute the inventory remaining from the previous generation until insufficient TRUs remained to fuel one transmutor for its lifetime. The TRUs left over at the end would require disposal with other HLW.

2b. The ALMR transmutor has been proposed to reduce the amount of TRUs in waste below the limiting amount that could be released to the environment according to 40CFR191, the EPA environmental protection standard for management and disposal of spent nuclear fuel, high-level and TRU wastes.[23] For ^{239}Pu, the most abundant radionuclide to be transmuted, the corresponding amount in the waste would be less than 0.03% of that in LWR spent fuel. As a result of multiple recycle in the transmutation systems, process losses to waste in each processing cycle would have to be less than about 0.007%. However, the ALMR/IRF project has not defined a development goal to attain such low process losses.

2c. During the entire life (ca. 30 years) of an ALMR transmutor, the amount of TRUs transmuted would be about half of the nearly constant inventory of TRUs in the reactor and its fuel cycle. To obtain further transmutation, the residual inventory would be transferred to subsequent generations of ALMR transmutors, as outlined in 2a. However, for ALMRs with a 0.65 breeding ratio and fueled with TRUs from LWRs (as proposed by DOE) the constant-power operating time required to reduce the inventory of residual TRUs to even 1% of the inventory of the referenced LWR once-through fuel cycle would be unrealistically long, on the order of many millennia (see 2b and Figure 4-3). The first century of constant-power transmutation could only reduce the inventory fraction to about 14%, too great to meet the ALMR's waste-disposal objective stated in 2b. More extensive transmutation would be required. A break-even, breeding ALMR would require even longer operation.

2d. In the declining nuclear power scenario outlined in 2a, the ALMR with a 0.65 breeding ratio could achieve a TRU "inventory fraction" as low as 9% in 100 years.[24] However, the lowest fraction attainable would be about 0.5%, corresponding to final operation of one module of a nine-module ALMR. The residual inventory would be too great to meet the ALMR's goal of waste reduction (see 2b) unless smaller terminal ALMRs could be justified.

2e. To achieve the same goals for meeting the EPA standard 40CFR191 as in 2b, the LWR transmutor could allow slightly greater process losses of plutonium and neptunium per cycle and would require slightly less time than the ALMR with a 0.65 breeding ratio. However, a lower per-cycle processing loss of americium and curium would be required. In the declining nuclear power scenario outlined in 2a, an S&T system using the LWR transmutor could achieve a similar inventory reduction in 100 years as that achieved using the ALMR of 0.65 breeding ratio (see 2d).

2f. Even though the ATW expects to achieve very low per-cycle process losses to waste, the frequent processing of the fluid fuel results in inventories of plutonium and neptunium in the ATW waste as high as 0.4% of that in the spent-fuel waste of the reference LWR once-through fuel cycle. For hypothetical constant-power operation over extended time it would take about 1,400 years for the inventories of TRUs in the aqueous ATW reactor to equal those in the ATW waste. Declining nuclear power operation (phase-out) would take about 90 years. For the nonaqueous ATW the corresponding times would be 350 years at constant power and about 50 years with declining power. This is shorter than for the ALMRs and LWR because of the very high fissile-specific power of the ATW concepts.

2g. The inventories of residual plutonium, neptunium, americium, and curium in the ATW reactor systems would require disposal in a geologic repository, as would other ATW waste such as Zircaloy pressure tubes from the aqueous ATW. It would be impossible for ATWs to effectively eliminate all long-lived radionuclides so that important residual radioactivity would persist "no longer than a human lifetime," as has been asserted by the ATW proponents.

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[23] Although the EPA 40CFR191 standard no longer applies to the proposed Yucca Mountain repository, it is possible that such release limits, or the equivalent thereof, could still become part of the new standard. Also, the ALMR/IFR project makes the assumption that EPA release limits applicable to LWR spent fuel could be used as such for HLW generated in transmutation.

[24] The "inventory fraction" is the ratio of the TRU inventory in the transmutor, fuel cycle, and waste to that in the spent fuel of a reference LWR once-through fuel cycle of the same electrical production.

2h. A plutonium-burning LWR is a simple and potentially useful transmutor system that could be implemented with present technology to decrease plutonium build up (it does not eliminate plutonium). Recovery of ^{14}C in PUREX reprocessing and qualification of new waste forms for separated ^{14}C and ^{129}I would still be required (see 1c). TRUs other than ^{239}Pu and ^{241}Pu would go to the waste. The times to reduce overall TRU inventories relative to the once-through LWR fuel cycle would be comparable to, but slightly shorter than, the times for the nonbreeding ALMR. During the first few hundred years of transmutor operation, the extent of TRU reduction would be little affected by the higher process losses of existing PUREX separations. Such reprocessing and the fabrication of recycle fuel would be simpler and somewhat less expensive than for an LWR transmutor with full recycle of all TRUs. However, reprocessing of even multiple-recycled plutonium, without recycling other radioelements, would reduce higher concentrations of transplutonics in discharge fuel and would make reprocessing difficult. At the present price of natural uranium, the cost of new U.S. facilities to reprocess LWR spent fuel and to fabricate recycled plutonium as MOX fuel would be prohibitive and the benefit to waste disposal doubtful.

2i. For a given electrical production capacity, some concepts for transmuting TRUs require less fresh uranium than the base-case LWR once-through fuel cycle. However, the transmutation systems that achieve the highest net consumption rate of TRUs also have a significant need for fissile make-up. Thus, their operation would still require an appreciable amount of fresh uranium. Developing and deploying TRU-burning ALMRs could lead eventually to ALMRs operating as self-sustained breeder reactors requiring no fresh uranium ore, but with much lower net consumption rate of TRUs.

3. The S&T systems differ widely in their state of technological maturity and present a broad spectrum of development issues, risks, costs, and schedules. The most mature system concept for transmuting TRUs, based on the use of LWRs, needs fuel-cycle development and would require about a decade and significant financial resources to reach the point of deployment. Compared with the LWR system, an ALMR/IFR system for transmuting TRUs would require more financial resources and take longer (perhaps a decade and a half) to reach the point of deployment. Beyond this, the ATW concepts would require major development before even the chances of success can be realistically assessed. An LWR- or ALMR-based system would require less development for transmuting only the plutonium from reprocessed spent fuel than for full TRU recycle.

3a. The most mature system approach would use the LWR together with aqueous reprocessing. Current LWRs would be suitable although advanced LWRs could offer cost and safety advantages when licensed. This thermal reactor system has the shortest time and lowest cost to complete development and full-scale demonstration of technical performance and system costs, about 8 to 10 years at a level of effort estimated at perhaps $50 to $100 million per year, exclusive of the cost of construction of any major test facilities. It also has the highest chance of successful development. Significant issues for reprocessing and for recycle fuel are posed by the multiple recycle of TRUs, in which higher MAs would tend to build up due to the LWR's thermal neutron spectrum. For transmuting actinides and selected fission products in addition to plutonium, an LWR system needs (1) further development, scale-up, and cost assessment of improved aqueous separations technology; (2) refinement of reactivity control for operation with TRUs; and (3) technology and procedures for licensable quality assurance and performance verification of refabricated recycle fuel containing significant amounts of MAs. For recycle of plutonium only, the required development in the three areas would be significantly simplified and could be accomplished sooner at lower cost than for full TRU recycle.

3b. The ALMR has been under development for decades as a fast breeder reactor based on plutonium recycle. However, it could be modified to operate as a TRU transmutor. No transmutation of fission products is envisaged with an ALMR. For burning TRUs, DOE proposes an ALMR/IFR with integral pyroprocessing of its spent fuel, which has not yet been demonstrated at pilot scale. The EBR-II fast reactor operating with a breeding ratio near unity has demonstrated a capability for passive safe shutdown. However, considerable experimental and calculational effort at breeding ratios of 0.65 or lower would be necessary to develop a database to support licensing with a reactor core that is fueled for full recycle (as proposed by the ALMR/IFR project) and contains higher-mass TRUs and fission products such as the lanthanides. However, the fast neutron spectrum of the ALMR would generate much fewer higher-mass actinides than the thermal reactors. Compared with an LWR and an ATW, an ALMR/IFR has an intermediate time and cost to complete development and full-scale demonstration of technical performance and system costs (about 15 to 20 years at a level of effort estimated at perhaps $100 to $150 million per year, exclusive of the cost of construction of any major test facilities) and an intermediate chance of successful development. Whether for full TRU recycle or plutonium-only recycle from spent LWR fuel, a low threshold cost for spent LWR fuel reprocessing is required for a viable ALMR/IFR burner system. Thus, its chance of successful development is lower than that of an ALMR system optimized for power production.

3c. The ATW concepts employ a subcritical thermal reactor using fluid fuel with integrated reprocessing. The proposals have high performance objectives for burning TRUs and key fission products. The ATW systems pose major engineering and materials challenges due to the extraordinary operating conditions in the reactor and beam target. The high power proton accelerator, which provides spallation neutrons for multiplication by the subcritical assembly, may be the best understood part of the system. Heat removal is a major issue for development as well as for safety and licensing. Also, the possibility of reactor transients, unevaluated in any detail at present, may require means of reactivity control beyond that afforded by merely turning off the accelerator. Moreover, the concepts use fluid fuels that involve unproved technologies for fuel fabrication and reprocessing, raising a host of operational and safety issues. The onsite reprocessing system has unique requirements for high reliability and maintainability, but a detailed concept has not yet been completed. The high thermal flux of the ATW would produce the greatest fraction of higher mass actinides of any of the principal concepts evaluated. Indeed, the high level of alpha and neutron radioactivity during onsite reprocessing and recycle-fuel circulation poses severe problems for the aqueous-based ATW-1 concept. For all the ATW concepts, the overall system economics are uncertain and are more sensitive than the other primary S&T concepts to the economics of feed material from reprocessing LWR spent fuel. Thus, the ATW concepts are in a qualitatively different position than the ALMR- and LWR-based concepts—extensive research and development would be required even to ascertain whether an ATW is technically feasible. For these reasons, the committee regards the ATW as a much less certain approach, with a questionable chance of successful development. If feasible, the ATW may require more than 20 years of high cost for development and full-scale demonstration of technical performance and system costs.

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Chapter Five

Defense Wastes

A long-recognized problem of the U.S. nuclear weapons program is the large amount of nuclear waste it has produced at several large sites across the nation. Defense wastes is a category that includes many types of nuclear waste of varying degrees of hazard. The range of defense wastes includes not only spent-fuel rods and contaminated equipment that are analogous to the wastes resulting from civilian light-water power reactors, although usually different chemically, but also a wide variety of waste types that have no counterpart in the present civilian program (e.g., the residues from fabricating plutonium components for nuclear weapons).

THE HANFORD TANKS

The Hanford site that produced weapons plutonium has the largest volume of wastes among U.S. sites and is widely believed to pose serious health and safety problems. Many of the conditions at Hanford can be found in varying degrees at the other material production and processing sites for nuclear weapons systems. A considerable amount of radioactivity remains on the site, more than half of which is in the form of aqueous solutions and solid residues in 177 underground mild steel tanks of high-level waste (HLW). A map of the Hanford site tanks and evaporators is shown in Figure 5-1; the 177 tanks are clustered in various locations within the "200 area" processing complex. The tanks are of two general types: 149 single-shell (SS) tanks constructed over the period 1943 to 1964 and 28 double-shell (DS) tanks built between 1968 and 1986. The latter have two steel walls with a void space between that provides leak detectability and additional containment in the event of a leak of the primary shell. The void space also provides access that permits some nonintrusive monitoring of the tank contents. A cutaway view of a typical SS tank is shown in Figure 5-2. The older SS storage tanks are well beyond their design lives, and 67 of them are assumed to be leaking, which resulted in draining and evaporation of much of the water from the solutions stored in them. The more recently built DS tanks are still sound and none have been found to leak.

Apart from the tank wastes, the second largest source of radioactivity at the Hanford site is the encapsulated radioactive sources. These consist of two types: sealed capsules of ^{90}Sr and ^{137}Cs, and unprocessed irradiated reactor slugs used as fuels from an onsite reactor (the N-reactor). The sealed strontium and cesium capsules are the result of a concentration process in which part of the liquid contents of the tanks was processed and the effluents returned to the tanks. The capsules are in sealed containers stored in deep water-filled pools (basins) that cool and shield the radioactive materials. The slugs are also stored in basins, but some are not well contained. Some additional radioactive residues at Hanford are contained in the form of locally contaminated soils used as waste disposal pits (cribs) over many years of low-level waste (LLW) disposal activities at the site.

This chapter deals only with the more contentious issues surrounding the tank wastes. It examines separations options that could be considered for remediation of these wastes and is meant to be suggestive of issues and solutions to be considered for the clean-up of other defense high-level wastes.

The 66-million-gallon total volume of waste in the Hanford tanks, and the debate over the appropriate treatment for its conversion to a suitable permanent waste form, make Hanford tank waste a primary focus of current DOE remediation efforts. A major concern about the Hanford tanks has been the possibility that chemical instability of the contents of some tanks could conceivably lead to uncontrolled chemical (not nuclear) reactions, with consequent tank rupture and release of radioactivity into the environment. After several years of evaluation and study, there is growing consensus that the likelihood of chemical instability is much less probable than initially believed. As a result, the emphasis is shifting from mitigation—elimination of safety hazards, such as the slight possibility of disruptive chemical reactions—to remediation, the resolution of underlying problems via the removal, separation, and disposal of the radioactive materials in safely storable waste forms.

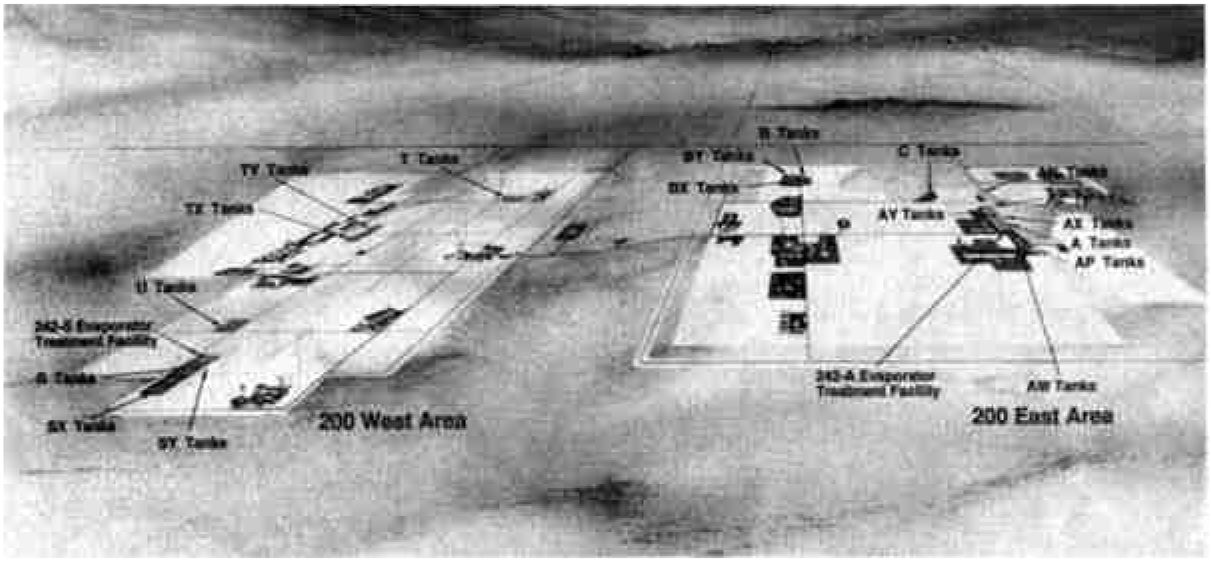
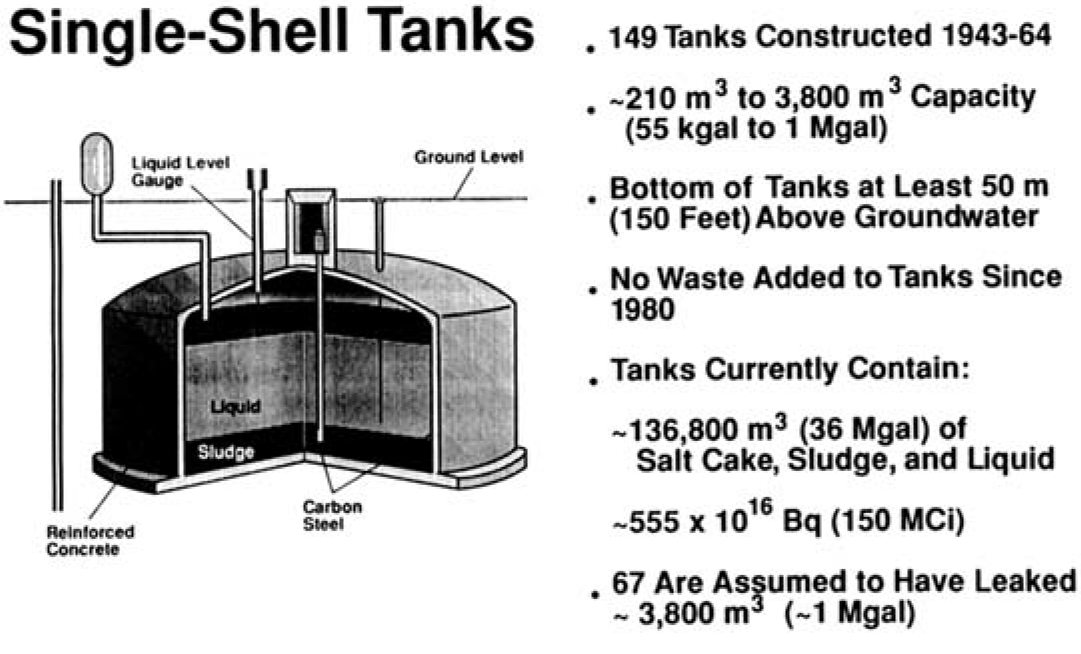


FIGURE 5-1 Hanford site tanks and evaporators.



Single-Shell Tanks

- 149 Tanks Constructed 1943-64
- ~210 m^3 to 3,800 m^3 Capacity (55 kgal to 1 Mgal)
- Bottom of Tanks at Least 50 m (150 Feet) Above Groundwater
- No Waste Added to Tanks Since 1980
- Tanks Currently Contain:

  ~136,800 m^3 (36 Mgal) of Salt Cake, Sludge, and Liquid

  ~555 x 10^{16} Bq (150 MCi)
- 67 Are Assumed to Have Leaked ~ 3,800 m^3 (~1 Mgal)

FIGURE 5-2 A cutaway view of a typical single-shell tank.

The Hanford tank wastes pose a challenge for separations (and possible transmutation), the study of which may supply useful information for the remediation of other defense sites and wastes. The development and successful execution of procedures for remediation of the Hanford site would be perceived as major progress for the whole defense waste activity and could serve as models for clean-up efforts at the other defense sites in the United States, including the Feed Materials Processing Center (Fernald), Y-12 Plant, Rocky Flats Plant, Mound Laboratory, K-25 Plant, Los Alamos, and the Savannah River Site, among others. (A summary of recent activity and plans at the Savannah River Site is presented in Appendix E.) All of these sites can be remediated, except probably contaminated soils, with known techniques; however, further development of the techniques is desirable to achieve optimum clean-up at a minimum cost in conformity with remediation standards.

Inventory and Characterization of Stored Waste

The combined waste volume in all Hanford tanks is estimated to be 250,000 cubic meters (66 million gallons), of which about 190,000 cubic meters (50 million gallons) are contained in the SS tanks. These hold approximately 155 megacuries (MCi) of radioactivity, while the DS tanks contain 110 MCi. A summary of the chemical and radionuclide inventory of the tanks obtained from records of past operational data is presented in Table 5-1. At least 99% of the current radioactivity (in curies) is due to ^{137}Cs and ^{90}Sr, even though during the 1967–1984 period, approximately half of the quantities of these fission products in the SS tanks were removed and placed in capsules. Because the Hanford tanks were made from carbon steel rather than stainless steel, the wastes were made strongly alkaline with sodium hydroxide to minimize corrosion, resulting in the high current content of sodium nitrate and sodium nitrite salts in the tanks. There is also a waste component known as "organic complexant concentrate," which was generated during the processing of many types of onsite residue fractions, as well as some offsite residues. In the DS tanks, this variable-composition fraction constitutes about one-third of the waste volume; the balance comes from both ion-exchange processing and tributyl phosphate (TBP) extractions that were routine production operations at Hanford's Plutonium Finishing Plant.

The composition of the material in the Hanford tanks is heterogeneous in all phases, both within a given tank and also among different tanks. A typical tank contains insoluble residues plus solid excess salt, precipitated residues (sludges) at the bottom of the tank, an intermediate layer of residual liquid that is saturated with a variety of salts and contains salt crystals in suspension, and a crust of low-density salts floating on the central liquid layer. (The extent of horizontal heterogeneity within each layer is unknown but can be expected to increase with the viscosity of the tank contents.) These waste materials have been generated over the past 50 years by three different chemical processes (described in Chapter 3) used to recover plutonium from irradiated uranium. Some of the differences in tank composition can be attributed to a change in the target cladding used for the reactor fuel charges, i.e., the change from the aluminum jackets used in the earlier reactors to the zircalloy cladding used for the N-reactor. Two other reasons are the recycling of the wastes from early bismuth phosphate plutonium recovery and purification processes to recover the uranium content, and the Irradiation Source Program that recovered ^{137}Cs and ^{90}Sr from much of the old waste to make intense gamma irradiation sources for industrial uses (such as sterilization of medical supplies and preservation of food for the military).

As mentioned earlier, the total inventory (Table 5-1) of significant species in the Hanford underground tanks is fairly well known, although the specific contents of some tanks and their layers are not. The sludges in the SS tanks are known to contain substantial amounts of mineral-like nonradioactive aluminosilicate solids known as cancrinites, presumably formed by reactions of aluminum salts with various silicate residues. Similarly, a significant portion of the insoluble residues in the DS tanks consists of hydrated zirconium oxides, hydroxides, and complex precipitates containing zirconium, silicon, aluminum, iron, and/or other minor elements. The uranium and transuranic (TRU) elements found in the tank wastes typically are precipitated under conditions such as those existing in the tanks, and some radionuclides may have been incorporated into the complex chemical structure of these mineral-like residues formed from nonradioactive elements. The amount of individual radioactive fission products present in the total contents of all the tanks and cribs can be estimated from historical operational power levels of the reactors and the decay-rate constants for each isotopic species. The bulk amount of salts in the tanks can be estimated from records of the volume and composition of acid solution waste discharged to the tanks and the amount of sodium hydroxide added to neutralize the excess acid.

The Hanford site's program for core analysis of the tanks has been driven by regulatory and safety concerns. Continued experimental and modeling studies increasingly indicate that there is an extremely low probability of encountering serious chemical instability problems even in those tanks of special concern, i.e., the tanks emitting radiolytic hydrogen and other decomposition products in significant quantities on a periodic basis and those containing potentially energetic compounds in contact with oxidizing salts (the organic compounds and ferricyanide-nitrate salt reaction issue). While sampling of the gas phase above the residues and analysis of one or two cores of residues per tank is useful to satisfy questions relating to possible safety issues, it is of little value in designing chemical remediation processes, particularly if the horizontal heterogeneity is extensive. The information obtained by this core sampling will

TABLE 5-1 Summary of the Chemical and Radiochemical Inventory of the Hanford Tanks

Material Type	Double-Shell Tank Waste	Single-Shell Tank Waste	Contaminated Soil	Total
Total Volume (m^3)	7.8E+04	1.4E+05	1.0E+05	4.9E+05
Density (g/cc)	1.5	1.6	1.6	1.9
Total Mass (metric tons)	1.2E+05	2.2E+05	1.6E+05	9.2E+05
Chemical Constituents (kg)				
Ag	4.4E+02			4.4E+02
Al	2.2E+06	2.2E+06	2.1E+0S	4.6E+06
As	8.4E+02			8.4E+02
B	2.6E+03			2.6E+03
Ba	1.8E+03			1.8E+03
Be	9.9E+00			9.9E+00
Bi		2.6E+05		2.6E+05
Ca	1.7E+04	1.3E+05		1.5E+05
Cd	6.7E+03	4.0E+03		1.1E+04
Ce	3.8E-02	2.3E+05		2.3E+05
Co	8.1E+02			8.1E+02
Cr	6.8E+04	9.6E+04		1.6E+0S
Cs	3.9E+02			3.9E+02
Cu	9.8E+02			9.8E+02
Fe	1.0E+05	6.3E+05		7.3E+05
Hg	2.8E+02	1.0E+03		1.3E+03
K	1.4E+06			1.4E+06
Mg	5.4E+03			5.4E+03
Mn	1.4E+04	1.2E+0S		1.3E+0S
Mo	8.4E+03			8.4E+03
NH_3				0.0E+00
Na	1.7E+07	5.1E+07	1.0E+06	6.9E+07
Nb	5.4E+00			5.4E+00
Ni	1.1E+04	1.8E+05		1.9E+05
Pb	3.7E+03			3.7E+03
Rare Earths	1.2E+04			1.2E+04
Rh	4.4E+02			4.4E+02
Ru	5.5E+02			5.5E+02
Sb	1.9E+03			1.9E+03
Se	1.4E+03			1.4E+03
Si	6.0E+04			6.0E+04
Sm	5.7E+00	2.5E+01	8.3E-03	3.0E+01
Sr	2.5E+02	3.6E+04		3.6E+04
Tc	1.4E+03	9.4E+02	1.8E+01	2.4E+03
Th	8.3E+02			8.3E+02
Ti	4.5E+02			4.5E+02
Zn	2.3E+03			2.3E+03
Zr	3.1E+05	2.5E+05		5.6E+05
Anions				
F	3.8E+05	8.1E+05	2.2E+03	1.2E+06
Cl	4.5E+05	4.0E+04		4.9E+05
CO_3	1.1E+06	1.7E+06	1.8E+04	2.8E+06
$Fe(CN)_6$	3.5E+03	3.2E+05		3.2E+0S
I	1.5E+02	1.4E+02	4.8E+00	2.9E+02

Material Type	Double-Shell Tank Waste	Single-Shell Tank Waste	Contaminated Soil	Total
NO$_2$	5.0E+06	4.8E+06	5.7E+05	1.0E+07
NO$_3$	1.4E+07	9.7E+07	7.9E+05	1.1E+06
PO$_4$	2.2E+05	8.8E+06	7.0E+04	9.0E+06
SO$_4$	3.8E+05	1.6E+06		2.0E+06
OH	1.0E+07	5.3E+06	1.4E+05	1.6E+07
Total Organic Carbon	6. 1E+05	2.0E+05	8.8E+04	9.0E+05
Cancrinite		2.7E+06		2.7E+06
Diatomaceous Earth		3.5E+05		3.5E+05
Portland Cernent/Concrete		5.7E+04		3.9E+08
Soil			1.6E+08	1.6E+08
Water	6.6E+07	4.5E+07	2.1E+06	1.1E+08
Selected Actinides (1cg)				
U	3.6E+04	1.4E+06	2.4E+03	1.4E+06
Np	4.2E+01	4.6E+01	1.6E-01	8.8E+01
Pu	1.6E+02	3.8E+02	2.6E-01	5.4E+02
Am	2.9E+01	1.4E+01	4.7E-02	4.2E+01
Cm	1.9E-02	8.6E-04	2.9E-07	2.0E-02
Radionuclides (Ci)	Approximate Decay Date: Jan. 1996			
^{3}H	5.3E+02			5.3E+02
^{14}C	2.3E+03	3.0E+03	1.0E+02	5.4E+03
^{60}Co	4.7E+03	9.4E+04		9.9E+04
^{63}Ni	2.4E+03	2.9E+05	9.9E+02	2.9E+05
^{79}Se	8.1E+02			8.1E+02
^{90}Sr	1.1E+07	5.0E+07	1.7E+04	6.2E+07
^{90}Y	1.1E+07	5.0E+07	1.7E+04	6.2E+07
^{93}Zr		4.3E+03	7.5E+00	4.3E+03
^{95}Zr	2.6E-04			2.6E-04
^{95}Nb	4.8E-04			4.8E-04
^{99}Tc	2.4E+04	1.6E+04	2.7E+02	4.0E+04
^{106}Rb	1.9E+04	7.4E-01	1.3E-02	1.9E+04
^{106}Ru	1.9E+04	7.4E-01	1.3E-02	1.9E+04
^{125}Sb	6.9E+04			8.9E+04
^{126}Sn	0.0E+00	5.7E+02	4.9E+00	5.7E+02
^{129}I	2.7E+01	2.4E+01	8.2E-01	5.2E+01
^{135}Cs		7.3E+01	1.9E+00	7.5E+01
^{137}Cs	3.0E+07	1.6E+07	4.2E+05	4.7E+07
^{137m}Ba	2.9E+07	1.SE+07	3.9E+05	4.4E+07
^{144}Ce	2.1E+03			2.1E+03
^{147}Pm	1.1E+06			1.1E+06
^{151}Sm	1.5E+05	6.5E+05	2.2E+02	8.0E+05
^{154}Eu	5.3E+04			5.3E+04
^{155}Eu	4.3E+04			4.3E+04
^{226}Ra		3.2E-15	1.1E-18	3.2E-15

provide only very limited information about each tank's total contents.

Material Type	Double-Shell Tank Waste	Single-Shell Tank Waste	Contaminated Soil	Total
^{235}U	7.0E-02	2.0E+01	3.4E-02	2.0E+01
^{238}U	6.6E-01	4.6E+02	7.9E-01	4.6E+02
^{237}Np	3.0E+01	3.2E+01	1.1 E-01	6.2E+01
^{238}Pu	4.2E+02	4.4E+02	3.0E-01	8.6E+02
^{239}Pu	9.1E+03	2.2E+04	1.SE+01	3.1E+04
^{240}Pu	2.6E+03	5.4E+03	3.7E+00	8.0E+03
^{241}Pu	5.0E+03	4.5E+04	3.1E+01	5.0E+04
^{241}Am	9.9E+04	4.6E+04	1.6E+02	1.SE+05
^{243}Am		1.9E+01	6.5E-02	1.9E+01
^{243}Cm				0.0E+00
^{244}Cm	1.5E+03	6.9E+01	2.4E-02	1.6E+03

[a] Includes tank structures.

NOTE: The absence of a component estimate in the above table should generally be taken to imply the unavailability of information on that component that could be referenced, rather than that the component is not present in waste materials.

SOURCE: Boomer et al. (1994)

Performing the sampling and the subsequent material analysis needed for tanks that might present safety hazards would be costly, especially if enough samples were taken to compensate for the heterogeneity in the tanks, and could result in additional radiation exposure to workers. An extensive program of sampling of individual tanks for reasons other than resolution of safety questions is unnecessary, and sampling should be minimized to the amount needed to provide data for satisfactory risk assessment.

Remediation Processing Considerations

The complexity of remediating the Hanford tank wastes results not only from the variety of wastes generated by different processing techniques and reagents, but also from the thermal heat arising from fission-product decay energy. It would not be technically prudent to begin the processing needed to produce a suitable permanent waste form before the large majority of short-lived fission product isotopes had decayed to either stable end-products or long-lived isotopes that emit little decay heat per unit time. The length of storage of the Hanford wastes to date has been technically beneficial.

Any plan for converting the wastes stored in the underground tanks to a suitable permanent waste form should take into consideration that the relatively small amount of radioactive material present is highly diluted with nonradioactive solid salts and large volumes of saturated aqueous brines. Both the precipitated salts and the brines contain suspended or dissolved ^{137}Cs, ^{99}Tc, and ^{90}Sr; the TRU elements, which represent only a few parts per million of the total tank wastes, are to be found (usually together with uranium) in the precipitated sludges that constitute the bottom layer of tank residues. The earlier decision to maintain a pH greater than 13 has exacerbated the precipitation of normally soluble salts and greatly increased the bulk of the solid material in the tanks.

In order to separate the TRU elements from the largely nonradioactive residues, the deposits must first be removed from the tanks. This may be accomplished by washing the residues with high-pressure hot water; some mechanical techniques may also be required for physically scraping the residue from the tank bottoms. (There are a number of pipes and protrusions for instrumentation that would complicate mechanical removal.) Depending on the degree of clean-up chosen as a goal—a key issue—chemical dissolution techniques involving fluoride containing strong mineral acids may ultimately be needed to remove radioactive materials to the maximum extent possible. Such treatment would certainly corrode the existing steel tank surfaces and might damage the tank structure or open old leaks. A major concern is the extent to which complete removal, whether by chemical or mechanical means, will reopen leaks and release radioactivity to the soil.

The inventory totals and distributions from the records are sufficient to permit the selection and development of a basic separations process, but it would be necessary to proceed with pilot-scale operation to obtain the detailed technical and operational information needed to design a suitable remediation process system. Planning for full-scale processing requires recovering representative samples of solutions, washings, and washed residues from the tanks for analysis. The residues could be dissolved in fluoride containing mineral acid (probably nitric acid), and the resulting clarified solutions and residual solids could be analyzed. This would provide the information for defining additional chemical processes for treating the tank sludges or the saturated solutions. A logical next step toward remediation processing would therefore be to demonstrate pilot-scale sludge washing operations on feedstocks from several typical tanks. This would provide a sufficiently representative sample of sludge residue, which could be analyzed chemically to provide the information needed to evaluate additional chemical process options for separating the actinide fraction from the sludges. The composition must be available for the major fractions of the feedstock to ensure the successful operation of any of the proposed processes. The selected process must be demonstrated with representative fractions of actual feed material before any type of large plant for remediation processing is constructed. Both the pilot plant and the production facility should operate on a blended feedstock to reduce the effect of variations in tank feed. While the production plant should be designed to accommodate variations, the preparation of blended feed solutions is expected to be a normal part of any process. Variations in both the washing techniques and the tank contents need to be considered, averaged, and analyzed to ensure a stable average input during normal operation of the separations plant.

SCENARIOS FOR HANFORD TANK REMEDIATION

Decision Factors

Options for the remediation of the Hanford tank wastes can be weighed in light of four primary factors: (1) risk (local, regional or national, and worker safety); (2) technical feasibility; (3) public acceptability; and (4) cost.

While the committee focused primarily on the technical feasibility of the various separations options, with some discussion of costs, it was recognized that public acceptability and perceived risk are closely linked. The actual risks cannot accurately be defined until the separation processes are further defined, but a general rule is that risk will increase with the degree of handling and processing.

Figure 5-3 shows alternative processing scenarios defined by how far one proceeds through the diagram. The choice of path at each juncture should be based on the four factors noted above, using information from literature reviews, research and development studies, and pilot-plant tests on the actual waste product resulting from the previous steps.

At its most basic level, separations processing of the Hanford tank waste will divide the waste into LLW and HLW streams. The present plan for the high-level stream involves vitrification and disposal in a geological repository, while the LLW may be disposed of by onsite burial or indefinite storage onsite.[1] A decision not to proceed with further separations at any of the junctures shown on Figure 5-3 would result in the approximate number of HLW canisters for geologic disposal shown at the right of the figure. Each decision to undertake further processing reduces further the number of canisters requiring disposal. The cost of geologic disposal obviously depends on both the number of canisters and the regulatory protocols that are adopted. Early regulations proposed for a repository at Yucca Mountain limit the amount of defense waste to 10% of the total inventory of 70,000 metric tons of heavy metal (MTHM) equivalent. The Yucca Mountain repository cost has been estimated at $22 billion, with an additional $1 billion for a repository capacity increase of 10%; the $22 billion includes development costs of $13 billion. These costs would be assessed as charges against canisters placed in the repository. Some of the charge would depend only on the total quantity of heavy metal equivalent, but other charges would reflect the expense of producing and handling each waste canister. Consequently, the potential savings in repository costs that could result from reducing the number of waste canisters via chemical separations is unclear and nonlinear.

The costs of producing a suitable waste form, presumably glass in canisters made in a vitrification plant, can be estimated more accurately. Appendix E shows estimated costs of $0.3 million per canister for production of 38,000 canisters over 30 years, compared to $1.8 million each if only 1,000 canisters are made. Discussions are currently under way that may lead to the use of larger, more economical canisters for Hanford HLW. Repository disposal costs must be added to these estimated production costs.

Alternative Scenarios

The alternative processing scenarios shown in Figure 5-3 involve trade-offs between the quantity of HLW that would require deep geologic disposal (with its attendant costs and risks) and the amount of processing that must be done (and its attendant costs and possible risks). The scenarios also vary in how much LLW would require disposal. These processing scenarios are discussed sequentially below, from least to most chemical processing.

In order to provide a complete description of the full range of processing options, this section includes alternatives that have varying degrees of practicality. Current DOE plans are focused on the sludge-washing option with the possibility of further minimal separations. In order to com

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[1] The current reference approach at Hartford is to vitrify low level waste and store the vitrified products onsite.

plete the list of processing options, two other scenarios (which are not indicated in Figure 5-3) are considered briefly as well. These two scenarios represent the extremes of virtually no treatment and of complete removal of all radioactivity from the entire site.

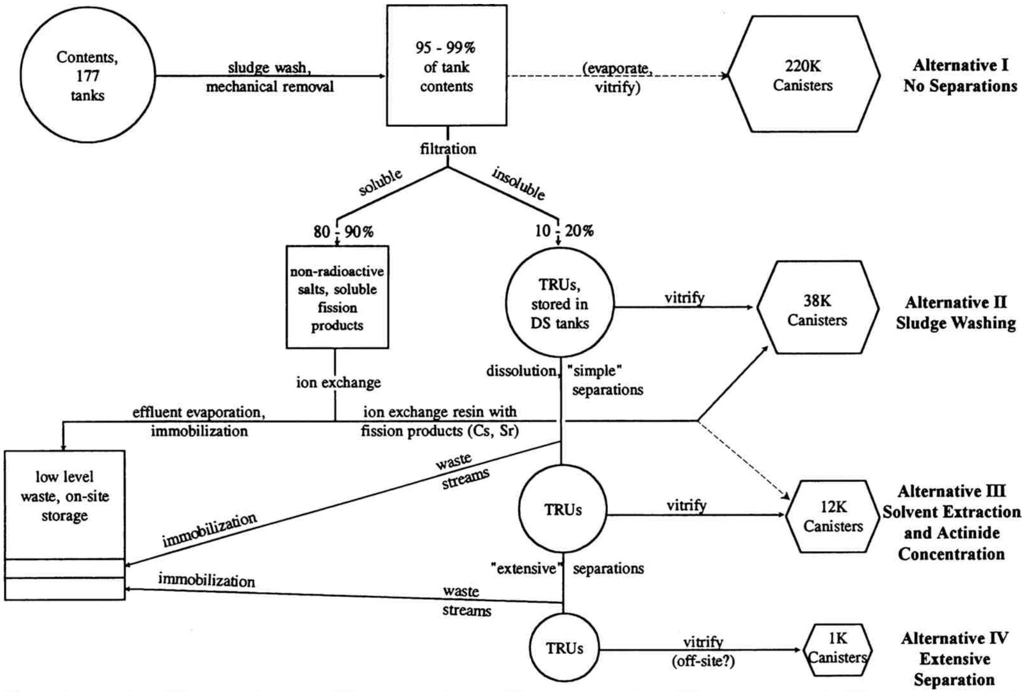

FIGURE 5-3 Alternative processing scenarios for Hanford tank wastes.

Risk estimates for the various scenarios are difficult to specify; to some extent, waste processing and repository storage merely reallocate the risk from the local Hanford environment to the locations affected by the transport and storage of the vitrified radioactive materials and to the workers involved in performing these actions. Total risk to the many people associated with the system may therefore increase.

No Separations Option

Physical removal of the tank contents by aqueous dissolution, evaporation of the water, calcination and recovery of nitric oxides, and vitrification of the residue has been estimated to result in approximately 220,000 canisters of mostly sodium aluminate glasses. Since the repository capacity is specified in tons of heavy metal equivalent, this quantity of canisters may not seriously affect the rules for eventual disposal in a geological repository (Johnson et al., 1993). However, their large number would surely exacerbate problems related to transporting canisters to the repository and to the repository's physical capacity, which in turn would present challenges to public acceptability. The diversity of feedstocks from the individual tanks would pose serious technical challenges for any type of vitrification plant, so blending of stocks would be essential. The cost of immobilizing and disposing of the high-and low-level waste fractions with this method (including construction of a vitrification plant, glass canister production over the plant's lifetime, and transportation and fees for geologic disposal) is unknown; a preliminary estimate—which seems very low—for the purposes of this discussion is approximately $15 billion (Johnson et al., 1993).

Sludge Washing

A treatment called sludge washing is a logical first step in any separations process that aims to reduce the volume of HLW requiring geologic disposal. In sludge washing, the saturated liquid is first pumped out of the tanks, and the remaining residue in the tanks is directly dissolved and washed with a hot, alkaline (pH > 10) solution. In most instances there will already be more than enough alkali present in the tanks to yield a pH of 10, and a strict policy of adding a minimum of reagents would be followed. After this washing, insoluble residues would be removed with remotely operated robotic systems, though the degree of removal is ill-defined at present. These hydraulic actions should remove most of the insoluble particulate matter from the tanks as a slurry, although an unknown quantity, perhaps 5%, of insoluble solid would probably remain. This residual material is expected to be intractable, and its removal by mechanical or chemical means could cause significant damage to the existing tank structures. If tank leakage resulted, it would increase releases to the environment.

The washing fluids and the soluble and suspended fraction pumped from the tanks should contain more than 90% of the sodium and cesium and 20-40% of the strontium; approximately half of the technetium would be expected to dissolve. The cesium may be more difficult than sodium to wash from the sludge. Removal of the strontium and cesium from these fractions could be accomplished by established separation processes such as zeolite adsorption and/or ion exchange. It may not be necessary to explicitly target the water-soluble portion of the technetium, because the ion exchange process proposed for removal of cesium and strontium from the aqueous solutions may also remove a large fraction of the technetium by adsorption into the selected exchanger. Technetium is rendered insoluble by reduction. Any remaining radionuclides in the solutions of sodium salts and wash fractions would be in low concentration.

The residue remaining after evaporation of the recycle wash water used in this solution-removal and salt-washing step would qualify as LLW but would probably require treatment by processes such as calcining or biological decomposition to decompose the nitrate and generate a satisfactory feed for the process used to produce the final LLW storage forms, which may be largely sodium aluminate. Environmental concerns relating to the calcining process could require recovery or decomposition of any nitrogen oxides produced during this step. In addition, calcining would produce effluent gases (from organic complexants and their decomposition products) that might require scrubbing.

The dissolution of the water-soluble fraction of the waste by sludge washing as described above would leave the insoluble sludge fraction, composed of particulate matter, insoluble portions of salt cake and the settled sludge. These materials should contain almost all of the alkaline-insoluble fission products, the remaining strontium, and, most important, essentially all of the actinide elements from thorium through curium. A large portion of the actinides in the insoluble fraction would be uranium compounds. These separated sludges could be stored in the available DS tanks until a processing decision is made for the next step.

The simple sludge-washing process (including zeolite treatment of the solution phase to remove cesium, technetium, and strontium, but without any further chemical separations among the water insolubles) could reduce the volume of waste requiring vitrification by an order of magnitude if the majority of the aluminum salts can be dissolved in the alkaline wash. The cost of sludge washing, removal of cesium, strontium, and technetium from the aqueous fraction, final evaporation of the water solvent, calcination of the residues, and onsite disposal of LLW has been estimated to be $5 to $10 billion (Boomer et al., 1994).

If the HLW resulting from this complete process—consisting of the adsorbed cesium, strontium, and technetium in zeolite and any remaining insoluble sludge removed from the tanks—were vitrified, it would result in approximately 38,000 canisters for geological disposal. The heat generated by the cesium and strontium would be the principal determinant of the number of canisters. The required capacity of the vitrification plant would be reduced relative to that needed for the "no separations option." It also should be possible to produce a feedstock with less compositional variation (and correspondingly fewer operation problems) than would be possible if the untreated tank contents were vitrified directly. Estimates of the total costs, including sludge washing, LLW disposal, construction of a vitrification plant, glass canister production over the plant's lifetime, and geologic disposal (including transportation and fees), are estimated at some $25 billion or more.[2] Costs may be lessened significantly if the aluminum salts are soluble in the caustic wash, resulting in less sludge to the glass ingots and therefore a smaller investment in vitrification to make the glass ingots.

The LLW resulting from this process would still contain trace levels of ^{137}Cs, ^{90}Sr, ^{99}Tc, ^{129}I, and other radionuclides. For this, concrete grout or a sodium glass may be a suitable (Class C) waste form for onsite disposal. It may be reasonable to consider using the emptied DS tanks for the permanent storage of this LLW, although this would not conform to current DOE agreements. The residues generated after the aqueous fraction has been evaporated to dryness, calcined, and converted to a permanent waste form (e.g., immobilized in glass or concrete) could conceivably be deposited in these tanks.

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[2] Boomer et al. (1994) gives a range of estimates from $18.27 to $24.87 billion for various sludge-washing options, to which the costs of tank-waste retrieval must be added. The latter is estimated at $5 to $15 billion depending on the aggressiveness of retrieval efforts.

Solvent Extraction for Actinide Concentration

Additional chemical processing has been proposed for the insoluble material that remains after the sludge washing to reduce further the number of HLW canisters requiring repository disposal. While complete dissolution of the concentrated sludge may not be readily achieved, treatment with strong mineral acids and hydrofluoric acid should take essentially all actinides into solution. The resulting solution could be subjected to one or more of the processes such as TRUEX described in Chapter 3 for solvent extraction of actinides. Such an actinide concentration step should further reduce the number of glass canisters of HLW needing geologic disposal, from 38,000 to approximately 11,000. The probability is high that this process could be technologically successful.

This actinide concentration approach is similar to the "solvent extraction (advanced separations)" case originally described by the Westinghouse Hanford Co., which includes separation of strontium and technetium (Johnson et al., 1993). Total costs for that option, including tank processing, LLW disposal, construction of a vitrification plant, glass canister production over the plant's lifetime, and geological disposal (including transportation and fees) were estimated in the range of $25 billion (Boomer et al., 1994). These numbers are very speculative; the cost of additional separations beyond sludge washing was estimated to be offset by the reduction in production and disposal costs associated with the lesser volume of HLW.

Extensive Separations

Further treatment of the non-TRU radioactive material discharged by TRUEX-like processes has been proposed to reduce the vitrification plant product to 1,000 canisters. The Westinghouse Hanford Co. estimated the cost of this option at $17.35 billion, to which retrieval costs of $5 to $15 billion must again be added (Johnson et al., 1993; Boomer et al., 1994). The combinations of new and existing separation processes proposed to achieve this further reduction in glass canisters was called the extensive separations proposal (Straalsund et al., 1992), and its block flowsheet is given in Figure 5-4. To be economically attractive, the costs of such further processing would have to be offset by the reduction in canister production and disposal expense, but the 10,000-canister reduction from the preceding option is estimated to save only $4 billion, based on the total per-canister costs estimated in Appendix E. It is very doubtful that the extensive separations process could be constructed and operated for $4 billion. None of the separation processes proposed for this option has been demonstrated beyond the laboratory scale, and some have not been tested even at that level. There would be major technological challenges to overcome, particularly as regards compatibility problems in the sequential separations systems required for this option as defined. In addition, the extensive separations option would also produce large secondary and tertiary waste streams that in turn would require concentration, processing, and disposal.

In-Place Option

The in-place scenario involves no removal of material from the tanks. The tank contents would be solidified in an insoluble matrix with a demonstrated, essentially permanent stability. Although this is the least expensive option, neither its risk nor its technical feasibility has been evaluated thoroughly. Current evidence indicates that this option would be unacceptable to the local public, although the low cost might be favored by the general public as other government costs increase. Using current technology, there is probably not enough space in the present tanks to stabilize all the soluble materials present to the degree that would be considered satisfactory. A few large new containers would have to be added on the site to hold the overflow as stable solids. The containment system would need to last well beyond the 300 years required for cesium and strontium radioactivity to decline to about 1/1,000 of the present level. The extremely long half-life problem of the TRUs and technetium would remain also.

Pristine Site Restoration

Restoration of the site to pristine condition is the extreme end of the remediation spectrum and would involve removal of all added radioactivity from every area of the entire site. This level of clean-up would require not only the removal of tank waste and other stored waste, but also the removal of the emptied tanks (which would still contain significant quantities of radioactive materials) from the site. It would also be necessary to remove the underlying sand, the contents and soil surrounding many other HLW disposal and spill locations (e.g., cribs and ponds), and to treat the groundwater as necessary to restore it to approximate the original environment. The costs of remediation at this level would be extraordinarily high. Since the entire site should be considered measurably radioactive, some rational standard for "low enough," or "as low as reasonably achievable" activity, would need to be adopted even to calculate the cost of this option.

The costs for pristine restoration have been estimated to be between $300 and $600 billion. Moreover, the level of chemical separations called for would be extremely high, with many attendant uncertainties, and a protracted period of research and development would be required to develop the necessary decontamination and restoration technologies.

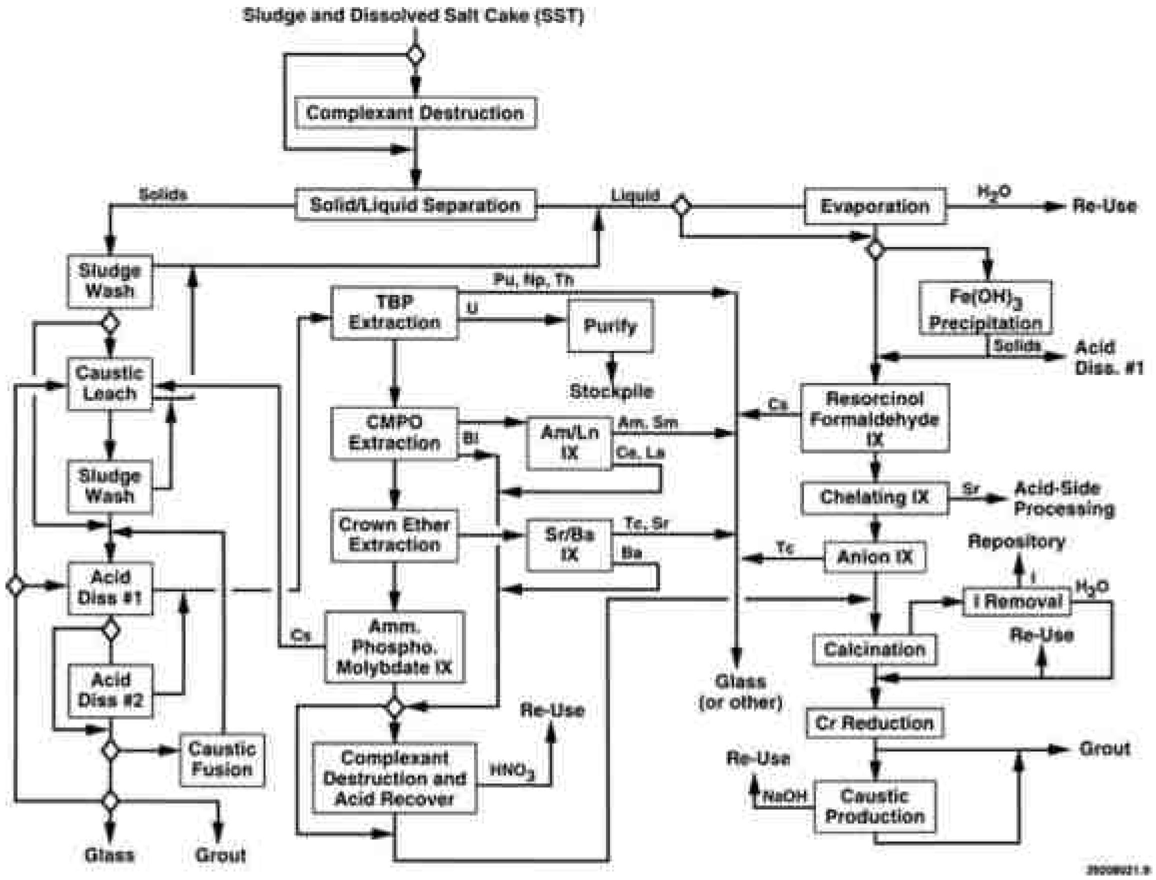

FIGURE 5-4 Flowsheet for the extensive separations option process.

Transporting the waste would also be an increased risk to all populations along the routes, adding to both costs and political opposition.

TRU Isolation for Transmutation

Producing the relatively pure target-element material required as feed to transmutation systems that would destroy the TRU content of the Hanford tanks (a quantity of less than one metric ton) to the degree claimed by the most optimistic proposals would require even more extensive separations and recovery than the options proposed above. Other transmutation approaches might be less costly (and less effective), but the reactor operations alone would be a large expense, and all transmutation systems require an elaborate reprocessing plant. Because the spent nuclear fuel scheduled for the first national repository contains over 600 metric tons of TRUs, there appears to be no justification for performing expensive further separation processes so that a fraction of the defense waste TRUs can be transmuted rather than sent to a repository. Both the IFR project scientists at Argonne and the ATW project scientists at Los Alamos agree that application of their transmutation systems to the Hanford tank TRUs may not be justifiable because the small quantity of actinide isotopes involved even in the relatively impure state that would be recovered from a PUREX-type separation could be disposed of through any of the glassingot options. Therefore, the Hanford TRUs—and by extension those from all defense waste sites—can be disposed of with little additional strain on the proposed national repository in vitrified form at considerably less cost than that of any of the proposed transmutation options.

CONCLUSIONS AND RECOMMENDATIONS

The Hanford storage tanks contain the defense nuclear waste of principal concern at present. Wastes stored at Savannah River, Idaho Falls, Oak Ridge, and elsewhere present serious problems, but they are smaller in total volume and present somewhat less urgent issues by comparison with the Hanford tank wastes. The primary goals of tank treatment are to mitigate safety concerns and to remove the ^{137}Cs and ^{90}Sr, which are the major sources of radioactivity in the tanks, thus reducing the amount of HLW for disposal.

Separations of the Hanford tank wastes would involve removal or destruction of sodium nitrate and sodium nitrite salts added during processing. These salts (along with aluminum salts, sodium carbonate, and sodium hydroxide) comprise the bulk of the waste. The simplest option for tank-waste remediation is sludge washing with salt dissolution to remove the bulk of nonradioactive salts, ^{137}Cs, and part of the ^{90} Sr from the tanks, reducing the volume of HLW by an order of magnitude. Removal of ^{137}Cs and ^{90}Sr from these bulk waste constituents would produce a LLW that is approximately equivalent to the Nuclear Regulatory Commission Class C waste category. This is the option currently being pursued at Hanford.

The committee makes the following recommendations relative to the wastes stored in the Hanford tanks:

1. Dedicated transmutation systems should not be considered an option for the remediation of the Hanford tank wastes, or of the other weapon system reprocessing wastes that exist in lesser amounts and analogous kinds at Savannah River, Idaho Falls, and Oak Ridge. The major problems at all these sites are the cesium and strontium fission products which are not candidates for transmutation. The other radioactive isotopes are small in quantity and can be managed safely by other disposal options.
2. Core sampling at Hanford should be employed principally for those tanks for which chemical instabilities are a special safety concern. Extensive sampling is not required for creating a remediation processing plan, and it would increase the likelihood of worker exposure.
3. Highest priority should be given to pilot-scale demonstration of the process involving dissolution of water-soluble sludge and washing of water-insoluble sludge. This involves recovery of cesium, strontium, and technetium from the wash solutions, storage of the insoluble residues, and disposal of the remaining salts from the solutions as LLW.
4. Actual concentrated residues from the sludge dissolution and washing processes in the recommended pilot plant should be used in development studies to determine whether further processing of the HLW is technically and economically justifiable.
5. Because of uncertainties associated with all the processing options, even with the minimum sludge wash treatment, the types and volumes of HLW requiring vitrification cannot yet be defined. Consequently, construction of a Hanford vitrification plant should ensure that the plant design will be compatible with the type and volume of waste requiring vitrification.
6. The option of pristine site restoration appears to be highly impractical technically.
7. A carefully coordinated national research and development program should be instituted to evaluate chemical separation options for the clean-up of DOE defense sites and residues, with emphasis on technology development and demonstration of candidate processes. This program should focus on demonstrating economically viable, practical processes for application to current problem areas.

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Chapter Six

Analysis of the Issues

IMPACT OF S&T ON WASTE REPOSITORY

All of the proposed Separation and Transmutation (S&T) systems have the potential to affect the design and long-term performance of nuclear waste repositories. Since these systems would require reprocessing of spent fuel, they would allow the separation of transuranics (TRUs) and selected fission products from spent fuel. The waste going to repositories would have less thermal power, would contain a reduced quantity of certain isotopes, and could be incorporated in waste forms with good integrity. These separated species would then be recycled in an ongoing fuel cycle of additional reactors and maintained at constant inventory per reactor until ultimately reduced in quantity in a possible phase-out of nuclear power. The claims of the possible effects of S&T on the first repository in the United States have ranged from eliminating the need for a repository to improving performance so as to provide a sounder technical basis for licensing. It has been proposed further that reducing the decay heat of the waste would permit the repository's capacity to be increased, thereby eliminating or postponing the need for a second repository.

Evaluation of Possible Repository Releases

A performance assessment of nuclear waste repositories relies on evaluation of possible future conditions and events that might allow radioactivity to be released. Two general areas are typically addressed: (1) transport of radioactivity by fluid flow (water or air) into the human environment, and (2) disruptive events and inadvertent human intrusion that cause a portion of the repository contents to be directly transported to the surface or injected into the groundwater system.

A limited number of performance assessments have been performed for repositories in the United States, for example, evaluations for the Waste Isolation Pilot Plant (WIPP) (Sandia National Laboratories, 1992). These assessments indicate its general compliance with the original EPA containment standard. For the WIPP, human intrusion would provide the primary mechanism for any appreciable radiation release to the environment. No direct assessments of the risk of radiation exposure to individuals have been made for human intrusion scenarios by current repository projects in the United States. They Yucca Mountain project has completed an initial set of calculations (Wilson et al., 1994) that will be further refined as data are obtained from site characterizations. The calculations show that the probabilistic estimates of release are within the limits of the EPA standard and also that the primary concern is the gaseous transport of ^{14}C to the surface. A larger set of calculations that have been performed evaluate the long-term release of radioactivity as well as the consequence and risk of individual exposure for other repository settings. Many of the concepts relating repository performance to transmutation have been addressed in the report by Ramspott et al. (1992).

In general, all calculations made to date, regardless of the basis for the evaluation (i.e., release to the environment or individual dose) indicate the following:

- The release of radioactivity to the environment via groundwater pathways is dominated by soluble, nonsorbing, long-lived radionuclides.
- The dominant products released via human intrusion or disruptive events, which can occur earlier in the life of the repository and extract radioactivity directly from the waste package, would be TRUs, such as plutonium and americium.
- In the case of Yucca Mountain, gaseous transport of

^{14}C could be the major contributor to predicted releases above the limits of the original EPA standard.[1]

Effects of S&T Processes

The S&T concepts with significant thermal-flux levels (accelerator transmutation of waste [ATW] and light-water reactors [LWR]) have the potential to reduce the mass of fission products in the reactors and repository wastes. In addition, the processes consume fissionable TRUs as fuel. Consequently, these concepts would reduce the risks associated with releases due to both human intrusion and fluid transport provided no additional fission products from external sources are laded into the repository. In the LWR case, reprocessing of spent fuel and transmutation of plutonium through recycling would ultimately yield increasing amounts of americium and curium. Their ultimate disposition would limit S&T benefits related to human intrusion releases.

The fast fluxes in the advanced liquid-metal reactor systems would primarily affect the magnitude of and consequently, the hazards related to the TRUs. Since the separated TRUs would remain in the operating fuel cycle, potential releases from human intrusion could be reduced. Fission products would not be reduced significantly, and fluid-transport release would not be directly affected. However, the advanced liquid-metal reactor (ALMR) program emphasizes the benefit of reduced heat-generation risks per unit of electrical energy predicted, allowing more fission products to be loaded into the repository. This greater concentration would eventually increase the figure for predicted individual dose related to aqueous-transport release.

The ultimate composition and character of the waste delivered to the repository will be determined both by the types of transmutation devices and by the reprocessing separations and vitrification operations—more americium and curium and less iodine and technitium will result from a thermal spectrum, for example, from transmutation. All S&T concepts provide the potential for reducing the quantity of some radionuclides in the waste product, thereby allowing it to be retained on the earth's surface. Reprocessing and inclusion of the products in a higher-integrity waste form without transmutation would allow some advantages in reducing the adverse impacts of the source term for both human intrusion and fluid-transport scenarios. Reprocessing also provides a significant advantage in that it can separate much of the ^{14}C from spent fuel. This allows the ^{14}C to be released to the atmosphere within current operational standards, stored on the surface for long periods, or converted to a nongaseous form for geologic disposal. While such dispositions would result in a marked improvement in predicted performance for Yucca Mountain, ^{14}C is probably best addressed by more careful examination to provide consistent disposal standards. It would seem unreasonable at this point to allow the performance of repository to be based on a long-term release of ^{14}C over thousands of years, when by simply addressing a different set of operating release standards, the same amount of ^{14}C could be released into the atmosphere without violating current regulations.

Finally, all S&T concepts will yield reprocessing products and secondary waste streams that could easily contain sufficient radioactivity to require disposal in a repository. It is unlikely that either the primary or secondary waste products could be kept below the 100-nCi/gm level for TRU wastes, which is the threshold level that differentiates TRU waste from low-level waste.

Elimination of all TRUs from HLW would reduce heat generation relative to spent fuel. At 10 years after discharge the TRUs in the spent fuel contribute 20% of the heat generation rate at that time, 60% at 100 years, and 99% at 300 years.[2] These percentages are also the percentage reduction in thermal power at those times if TRUs are all removed (see Appendix G for details). If this reduction in thermal power is coupled with a sequential waste emplacement approach, significant gains in repository capacity (4 to 5 times current capacity) could be achieved. (This equivalent would, however, eventually increase the predicted individual dose from aqueous transport from the first repository.) It should be noted that the current limit for Yucca Mountain (70,000 MTU) is less than the discharge expected from the existing LWRs (approximately 90,000 MTU) during their lifetimes. Yucca Mountain's capacity could be increased beyond the current level by one of the following: (1) using higher-density emplacement schemes or (2) more fully utilizing planned underground areas. In considering a thermal design strategy for the Yucca Mountain repository the characteristics of the unsaturated zone and the long-term heat generation by actinides in spent fuel must be considered. Thus a thermal design strategy for repositories in other rock types with other waste forms could be substantially different.

The wastes for permanent disposal will have varying concentrations of fissile materials depending on what process generates them. In most cases, the dominant fissile species will be ^{239}Pu and ^{235}U. Under some conditions, it may be possible for the waste deposit to reach criticality.[3]

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[1] In general, ^{14}C will, if the principal transport is by water, influence the residual radiotoxicity if the pH of the formation is not alkaline (pH 9). In that case, CO_3 and HCO_3 migration will occur. In the case of alk aline formulation, presence of large $CaCO_3$ concentration (e.g., in clay) is a geological buffer against CO_3 and HCO_3 diffusion.

[2] The integrated heat release will depend on the type of waste (reprocessed waste). The impacts on the repository are different for different repository types beyond the Yucca Mountain repository with its characteristic unsaturated zone.

[3] The possibility of autocatalytic criticality with some energy release has been raised recently (Bowman et al., 1994).

Avoidance of criticality can usually be achieved by adjusting the composition of the waste, size, separation of the waste canisters, or the type of overpack used. Because criticality calculations depend on the specific characteristics of the repository and waste, this issue was not dealt with further in this study.

Impact on the First Repository

Full implementation of an S&T system would provide a durable waste form depleted in TRUs and/or fission products (see Appendix G). Further, it could eliminate ^{14}C as a major contributor to predicted releases above EPA limits and could reduce heat loadings significantly after the second hundred years. Furthermore, the lower concentrations of key soluble radionuclides in the waste would result in reduced release to the environment over the long term.

Even though the goal of transmuting all waste to Class C or less (waste not requiring a repository) is a desirable goal, none of the concepts proposed is likely to achieve it. All would produce primary and secondary wastes in sufficient amounts to require geologic disposal. Extremely low process losses would be required, together with extremely selective separation processes, to result in wastes with TRU contamination less than the Class C requirement, 100 nCi/gm (100 nanocuries per gram = 10^{-7} Ci/gm).

The reduced thermal power could permit an increase in the initial loading density of the first repository and potentially in its capacity. This would have no direct effect on the need for first repository, although it would make a difference to at least the timing of and possibly the need for the second. Furthermore, thermal effects have not been shown to have detrimental impact on the long-term performance of a repository at Yucca Mountain. In fact, many have proposed that the elevated temperatures of the waste packages could provide for a "dry" period in which no water could interact with the waste packages, thereby reducing the potential dissolution and transport in the first thousand years or so. This topic is further discussed in Appendix G (Ramspott et al., 1992).

The S&T-released improvements in long-term performance would provide an advantage only if licensing and site characterization could be less extensive or lower in cost. No evidence is available to indicate that projections of long-term performance, which currently indicate relatively small individual and population doses, can simplify the licensing process or reduce the complexity of site characterization. The costs of construction and operation would not be significantly affected by incorporation of a high-integrity, lower-radioactivity-content waste form. The elimination of ^{14}C for a Yucca Mountain repository would provide a marked advantage for showing compliance with the original EPA standard, but the action required is unusually extensive and expensive, if done solely for this purpose. It is speculative to assume that release limits in the present standard will prevail in the study currently being undertaken by the National Academy of Sciences.

The net effect of the full implementation of any of the S&T systems proposed would not be to eliminate the need for a repository, but rather to offer the potential to improve its performance. Because of the time frame, to have any impact on the first repository would require a major and immediate commitment to an S&T system. If repository operations in the United States are to begin between 2010 and 2015, licensing application activities must be largely complete by 2001. At that time, the supporting technology necessary to obtain a license for the waste form in the repository environment must be available. In addition, full-scale reprocessing of LWR spent fuel must be licensed and available to support any of the transmutation systems. These operations must commence by about 2007 to furnish TRU to start ALMR by 2010. This is an extremely ambitious schedule unlikely to be achieved for any of the systems proposed, with the possible exception of LWRs.

Impact on a Second Repository

Implementation of an S&T system would have an impact on the second repository. Reduced heat load could allow for greater capacity in the first repository, provided legislative limits are removed. Its capacity could conceivably be expanded to more than twice the discharge of current LWRs or 200,000 MTU. This would be equivalent to an additional 40 or 50 years' discharge at current generation rates, thus postponing the need for another repository until about 2080. Similar benefits could also be provided by measures other than an S&T system, however. In the case of continuous electrical generation by systems with fairly low transmutation rates (ALMRs), TRUs could be continuously recycled to provide almost constant control of their inventory and material condition within the fuel cycle. Until that time when ALMRs are discontinued, requiring disposal of their inventory of TRUs in a geologic repository, only the fission products and secondary wastes in the resulting waste stream will need to be disposed. Of course, all the benefits of S&T for the first repository would also accrue to the second repository.

Conclusions

After considering the information above, the committee has reached the following conclusions about the impact of S&T on the repository.

1. There is no evidence that application of advanced S&T holds sufficient merit for the United States to delay

the development of the first nuclear waste repository to contain commercial spent fuel. Even with an S&T system, a geologic repository would still be needed.

2. Application of S&T does not hold sufficient merit to abandon the once-through fuel cycle.
3. While the need for a second repository could be delayed by S&T, there are several other ways, both legislative and technical, to increase the capacity of the first repository by a comparable amount.

TRANSPORTATION-RELATED ISSUES

Transportation of nuclear materials such as spent fuel and other wastes is the aspect of the nuclear fuel cycle that is the most visible to the public and touches them most closely. This section evaluates the impacts by S&T on nuclear material transportation.

Nuclear Material Transport

The potential impacts of nuclear material transportation increase with the toxicity and amount of nuclear material being transported and with the distance over which it is transported. These parameters, and thus the impacts, cannot be estimated for the various S&T options at present because many of the following factors have not yet been determined:

- the radionuclide composition of transmutation device fuels and wastes;
- the composition and volume of waste streams, both high-level and low-level, and
- the locations of the facilities that might be involved.

The current state of affairs and the various S&T options can be classified into three categories: once-through, onsite reprocessing, and offsite reprocessing. The generic transportation steps required by each of these categories are depicted in Figure 6-1. The committee used this framework, other information presented, and their expert judgment based on previous experience in nuclear fuel cycles to develop qualitative characterizations of the activity and volume for each of the various transportation steps. The results are shown in Table 6-1. It is clear from the above analysis that going from a once-through scenario to one involving onsite reprocessing of transmutation-device fuel would result in a significant increase in the amount of radioactive material being transported on highways and railroads because of the need to ship secondary wastes and the spent fuel, recycle fuel, and HLW from reprocessing. However, going from onsite reprocessing of transmutation-device fuel to offsite reprocessing would result in a comparatively small increase.

The characterization of the transportation impacts is of necessity severely limited. Because transportation of different materials can have very different risks per mile that overshadow differences in distances, the impact can be quite independent of the location of a facility. To achieve a representative figure, the transportation risk has been calculated for a case wherein LWRs are assumed to be located in Ohio (near the geographic centroid of LWRs in the United States), LWR reprocessing and ALMR plants are assumed to be located at Barnwell, South Carolina, and the repository at the Yucca Mountain study site in Nevada. Highway transportation was assumed. Incident-free and accident-related transportation risks to the public were calculated, as well as occupational exposures. The calculations, which used the ORIGEN-2 (Croff, 1980), HIGHWAY (Joy and Johnson, 1983), and RADTRAN 4 (Neuhauser and Kanipe, 1992) models, showed that the transportation risk was dominated by the transportation of spent fuel from the LWRs to reprocessing or the repository. The cancer fatality risk to the public was found to be 6.1×10^{-2} GWe/yr from the once-through cycle and 2.7×10^{-2} GWe/yr for actinide-burning in an ALMR with uranium recycling. These values, and even their relative magnitude, can be expected to change with the location of the various fuel-cycle facilities.

Transportation Package Technology

All three categories—once-through, onsite reprocessing, and offsite reprocessing—require one or more transportation packages (i.e., shipping casks and inserts) to move the radioactive material from one location to another. In most instances, the material being shipped is expected to have characteristics that do not differ substantially from materials now being transported. Spent fuels and HLW all emit substantial amounts of heat and penetrating gamma radiation that must be handled by the package. LLW and waste containing TRUs will be more voluminous, but their levels of radiation and heat emission will be much lower. The uranium product will also be voluminous, but will have low levels of radiation and essentially no heat emission. Packages currently exist for handling radioactive materials similar to all of those that would be generated in all the S&T scenarios. While package designs will depend on material-specific characteristics (e.g., physical dimensions, heat loads, and gamma-ray sources), there appear to be no technological impediments to their creation and certification.

There is one possible exception to the above. If a transmutation device with a thermal neutron spectrum (e.g., LWR, ATW, PBR) were to be used, substantial amounts of ^{242,244}Cm and ^{252}Cf would build up (see Chapter 4 for more extensive discussion). After repeated recycling, the amounts of these materials could increase to levels such that the neutron emissions would dominate the gamma rays, affecting the radiation shielding considerations (Croff et al., 1977; Alexander and Croff, 1980). If this becomes the case and offsite transportation is required, additional technology development will be required to design and build packages for highly neutron-active, heat-emitting spent fuels. These packages may be very large and expensive despite payloads that are small relative to those of conventional casks. The same packages will probably be required for the fresh transmutation-device fuel as well, because the neutron emitters are recycled with the fuel.

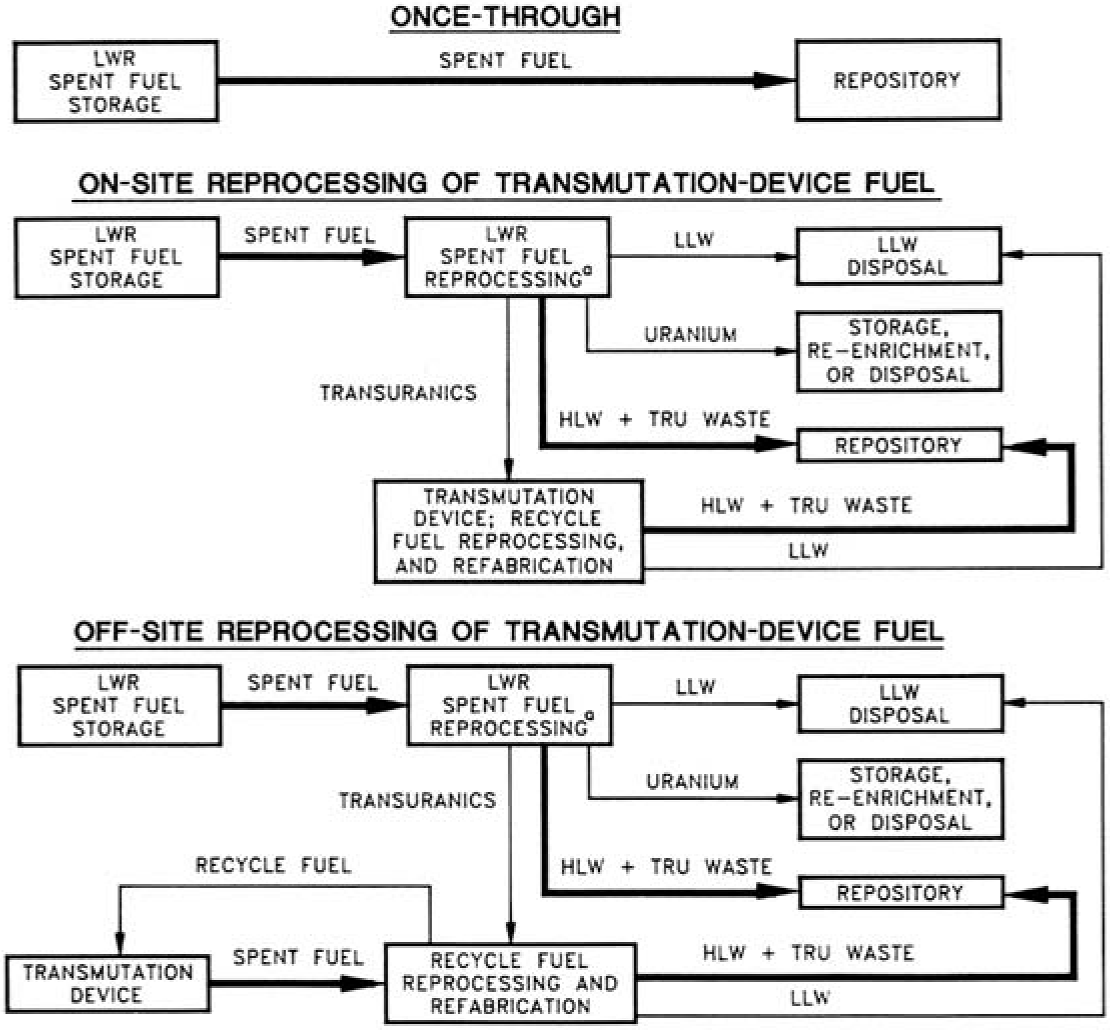

FIGURE 6-1 Transportation flows for once-through and recycle cases. NOTE: Bold lines indicate highest-risk transportation operations. [a] Could be colocated with recycle fuel reprocessing and refabrication.

Regulations Related to Transportation Packages

The U.S. Nuclear Regulatory Commission certifies (i.e., licenses) transportation packages under regulations it promulgates in Title 10, Part 71 of the Code of Federal Regulations (10CFR71). These regulations are based on design objectives (e.g., dose rates at certain distances, meeting various tests to assure structural integrity) and do not vary with the specifics of the material being considered. As a consequence, the regulatory structure required for the design and certification of radioactive material transportation packages for S&T scenarios is already in place, and it does not appear that significant modifications would be necessary.

Conclusions

The technology and regulations required to transport materials in an S&T fuel cycle are available unless extended recycle of actinides in thermal reactors were to occur.

TABLE 6-1 Characterization of Transportation for Once-Through and Reprocessing Cases

	Waste Characteristic		Category		
Transportation Step	Activity Level	Relative Volume	Once-Through LWR	Onsite Reprocess	Offsite Reprocess
LWR fuel from storage to reprocessing	H	M		X	X
TRUs from LWR to transmutation device[a]	M[b] -L[c]	L		X	X
LWR HLW/TRU waste or spent fuel to repository	H	M	X	X	X
LWR reprocessing LLW to disposal	M[d] -L[d]	H		X	X
LWR uranium to storage, reenrichment, or disposal	M[b] -L[c]	M		X	X
Transmutation-device spent fuel to reprocessing	H	M			X
Transmutation-device recycled fuel from reprocessing to transmutation device	M[d] -L[d]	M			X
Transmutation-device HLW/TRU waste from reprocessing to repository	H	M		X	X
Transmutation-device LLW from reprocessing to disposal	M-L	H		X	X

[a] Step not present when LWR fuel reprocessing collocated with transmutation-device spent fuel reprocessing.
[b] From pyrochemical processing.
[c] From aqueous processing.
[d] Collage of various waste streams of different activities.

PROLIFERATION ISSUES

There are three types of requirements—technical (knowledge and know-how), practical (facilities and personnel), and political—that a nation must meet to obtain materials in the quantities needed to make weapons (see Appendix H).[4] These requirements, like economic costs, pose barriers to nuclear proliferation. The following section, *Nuclear Nonproliferation and the Once-Through Fuel Cycle* , briefly reviews the way these barriers shaped nonproliferation policies and established the LWR once-through fuel cycle as the baseline. The section, *Proliferation Issues Related to Separations and Transmutation Systems* , uses that baseline to assess the proliferation issues raised by the proposed S&T fuel cycles. The committee's conclusions are summarized in *Summary and Conclusions*. Appendix H provides additional information and references on proliferation policies and safeguards and also assesses the proliferation aspects of four specific S&T concepts.

Nuclear Nonproliferation and the Once-Through Fuel Cycle

Each of the declared nuclear weapon nations—the United States, the United Kingdom, and the former Soviet Union, followed later by France and China—created large dedicated programs for the production of the fissionable weapon materials, as well as civil programs to develop nuclear technology for electric power production and medicine. As many nations began nuclear research and development programs, concerns about the "dual use" of nuclear technology led to the creation of an International Atomic Energy Agency (IAEA) in 1957 with dual responsibilities: to foster the peaceful uses of nuclear energy through technical assistance and to restrain the spread of nuclear weapons and the ability to make them.

The commercial LWR power system emerging in the 1960s answered the need for a nuclear fuel cycle that could enable many nations to use nuclear energy peacefully for medicine and power production while maintaining high barriers to nuclear proliferation (see Appendix H). Fresh LWR

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[4] The goals and criteria for materials accountancy are geared to the detection of quantities of safeguards significance of special nuclear materials for building a single nuclear explosive device, which the International Atomic Energy Agency takes to be 8 kg for plutonium, and, for uranium of > 20% enrichment an amount containing 25 kg of ^{235}U. Publicly available literature suggests that sophisticated designs to increase enutron efficiency and retard explosive disassembly could require significantly less fissionable material, e.g., perhaps 4.5 to 5 kg of plutonium and 15 kg of uranium enriched to approximately 95% (see Appendix H).

fuel contains low-enriched uranium (LEU) not usable for nuclear weapons. The technical and practical requirements for uranium enrichment put it beyond the reach of most nations for several decades. However, the spent LWR fuel poses a dilemma—the plutonium it contains is both bomb material and an energy resource. The intense radioactivity is a formidable hazard but would not prevent the reprocessing of the spent fuel and chemical separation of the plutonium.

To minimize the proliferation risk, the spent LWR fuel could remain in storage pools for many decades, and the issues of reprocessing or permanent disposal deferred, provided means could be found to ensure that the spent fuel would not be diverted and used for producing plutonium for nuclear weapons. That concern was addressed by a combined political-technical barrier, called international safeguards, instituted by the IAEA as an essential part of its responsibilities.[5] Thus, the LWR became the world standard for nuclear power and, as time passed, the once-through fuel cycle became the nonproliferation standard for the United States and many other nations.

The Nuclear Nonproliferation Treaty (NPT) in 1970 greatly enhanced the international basis for nonproliferation. Some 150 member nations are parties to the treaty, entailing obligations for both nuclear weapon and nonnuclear-weapon nations. Moreover, the basis of IAEA safeguards for NPT nations was considerably strengthened to achieve more uniform agreements with full-scope coverage (see Appendix H). However, several nations with sizable nuclear programs remain outside the NPT framework, although all of them have agreed to pre-NPT-type safeguards with the IAEA that cover some facilities, such as the price of technical assistance and the ability to import nuclear technology from the major suppliers.

The 1974 test of a nuclear explosive device by India, plus a series of public exposés in the 1980s involving Israel, Pakistan, and South Africa, revealed that those four non-NPT states had attained de facto but undeclared nuclear weapon capability using nuclear facilities not under safeguards (Spector and Smith, 1990). India and Israel used reactors and reprocessing to obtain plutonium. Pakistan and South Africa used enrichment to obtain High Enriched Uranium (HEU). Each public exposé was ambiguous enough to leave a figleaf of respectability for that nation's civil nuclear programs. Nonetheless, publicly available information summarized in Appendix H documents the fact that the weapon programs of the four nations, especially that of India, drew heavily on their civil nuclear programs.[6]

Still, this series of exposés did not seriously impugn the credibility of the IAEA safeguards system in effect, with a growing number of NPT signatories. Indeed, the gradual improvement in safeguards technologies, developed in cooperative programs between the IAEA and the nuclear powers and gradually deployed in NPT nonnuclear-weapon states, encouraged the perception that safeguards in NPT nations were adequate. From 1974 to 1980, U.S. nonproliferation policy aimed broadly at discouraging the civil use of plutonium and limiting the capability for enrichment and reprocessing to the European nuclear suppliers, the then Soviet Union, and Japan. During the 1980s, U.S. nonproliferation policy shifted to an emphasis on dealing with "rogue" nations, as several developing nations attained capability for enrichment or reprocessing on a production scale.

The Achilles' heel of safeguards—undeclared facilities and activities—made headlines in 1991 as a result of IAEA special inspections in Iraq after the Persian Gulf War. An NPT signatory since the 1970s, Iraq was discovered to be pursuing a covert nuclear weapon program in violation of its treaty obligations. Benefiting from intelligence information and acting under U.N. Security Council mandate, the IAEA inspection team discovered an extensive uranium enrichment program that had gone undetected in previous regular inspections (see Appendix H). Their discoveries showed that Iraq was perhaps 3 to 5 years from having a source of HEU sufficient to manufacture about one nuclear weapon a year (Fainberg, 1993). The discoveries also underscored the importance of IAEA's performing special inspections, as the situation arises, to ensure the credibility of its safeguards. Whether this precedent can be applied to other nations remains to be seen.

Recently, suspicious activities in North Korea, involving an undeclared reprocessing facility reported near Yongbyon, have raised apprehension in South Korea, Japan, and the United States. North Korea joined the NPT in the mid-1980s but balked at implementing a full-scope safeguards agreement with the IAEA. There is grave concern about the possibility that North Korea may have reprocessed fuel from a reactor that was shut down for about 100 days in 1989. At present, the political stand-off with North Korea and the fate of the IAEA safeguards there remain unresolved.[7]

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[5] Safeguard concepts are summarized in Appendix H. For a review of IAEA history and an extensive discussion of IAEA safeguards concepts, procedures, and problems, see Scheinman (1987) and Fischer and Szasz (1985).

[6] Two other developing states that are not NPT members, Argentina and Brazil, also developed the technology and facilities capable of "dual-purpose" uranium enrichment—Argentina using gaseous diffusion and Brazil using the centrifuge approach. In the late 1980s, the two nations ended their nuclear rivalry and entered into a stringent bilateral nuclear-safeguards regime backed up by full-scope IAEA safeguards (see Appendix H).

[7] On October 21, 1994, the long and difficult negotiations between the United States and North Korea bore fruit. The two nations signed an agreement under which North Korea would immediately freeze its nuclear weapons program and, over time, take steps to normalize its nuclear activities and facilities.

The spread of de facto but undeclared nuclear weapon capability underscores the gradual crumbling of the technical barrier to proliferation—over a 40-year period, the knowledge, know-how, and even the special equipment necessary to produce fissionable weapon materials have become available (see Appendix H). A major enrichment or reprocessing facility, however, remains a formidable undertaking. Some 5 to 10 years is required in most countries to construct and put it in operation. Thus, practical barriers to proliferation may still be effective if concerned nations show that they are willing to take timely action—political, economic, and, as a last resort, military—given information from satellites, overflights, inspections where allowed, and intelligence sources.

The U.S. "counter-proliferation" policy initiatives taken in 1993 give priority to bolstering the ability of the armed forces to respond to the proliferation of weapons of mass destruction in the post-Cold War world—particularly in the former Soviet bloc and in the hostile developing countries. It goes beyond political and economic efforts—meant to deter the acquisition of nuclear weapons or other weapons of mass destruction—to the potential use of military assets against a rogue nation or terrorist group with such weapons in their possession. Strengthened IAEA safeguards under a renewed NPT, perhaps buttressed by regional and bilateral agreements and safeguard regimes, could remain an essential mechanism to forestall the further spread of nuclear-weapon capability (Blix, 1992; Jennekens et al., 1992; Scheinman, 1992). This could be part of a larger initiative on cooperative security in the post-Cold War world, as recently proposed by two officials of the U.S. Department of Defense (Carter et al., 1992). Counter-proliferation means would remain a resort where cooperative means are not successful.

In September 1993, the Clinton administration stated its policy on nonproliferation and the use of plutonium: "… The United States does not encourage the civil use of plutonium and, accordingly, does not itself engage in plutonium reprocessing for either nuclear power or nuclear explosive purposes. The United States, however, will maintain its existing commitments regarding the use of plutonium in civil nuclear programs in Western Europe and Japan" (Office of the President, 1993). The policy states the U.S. intention to give nonproliferation greater priority in diplomacy and in consideration of regional security and economic matters, to seek to promote nonproliferation efforts, and to make nonproliferation an integral part of its relations with countries around the world. The policy reinforces the use of the LWR once-through fuel cycle and discourages any S&T undertaking that uses spent LWR fuel in the United States for the foreseeable future.[8]

Proliferation Issues Related to S&T Systems

The existence of major nuclear facilities and stores of fissionable materials that a nation has declared to be dedicated to civil purposes may indeed be perceived as benign by that nation's neighbors and other nations or, depending on the political situation, as a serious proliferation threat. The many possible proliferation scenarios can be classified into two main groups—*diversion* of materials from civil facilities or in transit, and *abrogation* of treaties or agreements by seizure of stores and facilities. Large stores of HEU for research, space power, or naval reactors, which could be further enriched to >90% ^{235}U, as well as stores of separated plutonium would be prime concerns. Large enrichment plants, spent-fuel reprocessing plants, and fresh-fuel fabrication plants are the subjects of particular anxiety because their operations entail the use of fissionable materials in bulk form, which is more difficult to safeguard than spent fuel. All the S&T proposals call for such facilities (see Chapter 4). Additional facilities of concern include large reactors, spent-fuel storage depots, and waste repositories, all of which are part of the LWR once-through baseline. The assessment below compares S&T systems to that baseline. Appendix H provides more detail on the proliferation implications of particular S&T technologies and evaluates four illustrative S&T systems.

Diversion Threats

One type of diversion scenario involves covert operations that result in the gradual removal of material in amounts that are small relative to the normal capacity of a material store or production plant. Another is an overt operation by a subnational or terrorist group to seize fissionable materials in transit, or even to attack a fuel-cycle facility in order to steal the materials. It may not be easy to distinguish either type of operation from one conducted by agents of the government itself, masquerading as a terrorist group. For most diversion scenarios, technological issues may be as important as political issues in assessing the risk.

National Diversion. The perceived risk of a national diversion of fissionable materials can be a serious matter—indeed, in 1981, Israel made an air strike against a reactor in Iraq just before it was due to start up. National diversion scenarios are potentially countered by stringent safeguards that apply materials accountability plus containment and surveillance techniques. At a typical reactor, the fuel rods provide a point of focus for safeguards (see Appendix H). The IAEA has devised special technologies plus a variety

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[8] S&T requires spent nuclear power reactor fuel reprocessing which has weapons proliferation implications and hence is of international concern. There would not be more plutonium around; in fact there would be less. But the plutonium in the spent fuel would be opened up and thus gets into a more accessible form.

of techniques to support inspections and checking of records at both regular intervals and randomly chosen times.

Accountability for bulk quantities of enriched uranium or plutonium, on the other hand, has its limitations under the best of circumstances. At present-day fuel reprocessing or fabrication plants, conventional materials accountability practices strive to achieve a materials balance on the order of 1% of the total fissile material in the facility. For example, a large reprocessing plant such as those in operation in Europe has an maximum throughput of about 100 MgHM of spent fuel per month. As is explained in Appendix H, this would yield perhaps 3 kg/d of weapons-grade plutonium; a 1% uncertainty could be equivalent to two plutonium weapons per year. This figure is high enough to raise concerns in many situations. Some proponents claim that a materials balance on the order of 0.3 to 0.1% could be achieved with advanced practices, however.

Typical present-day commercial facilities for aqueous reprocessing of spent fuel use semiremote maintenance with many maintenance portals and drains, which pose a challenge to secure containment and surveillance safeguards. However, the diversion potential might be reduced by designing plants for optimum containment and surveillance safeguards rather than for ready maintenance, e.g., with highly restricted access, entirely remote operation using robotics, and automated materials accountability. The potential for diversion might also be reduced by integrating the reactor and fuel processing facility, as described in the ALMR/IFR proposal (see Appendix H). Reducing the uncertainty level by an order of magnitude (to the 0.1% level) in facilities designed, constructed, and operated so as to facilitate the application of safeguards might be enough to remove national diversion as a serious threat associated with reprocessing and fuel fabrication. However, such a reduction would require much more cooperation with the IAEA on the part of facility designers, constructors, and operators than has typically been the case in the past.

Diversion by Terrorists or Subnational Groups. Seizure by terrorists or subnational groups can take many forms. Even if such groups have only limited means at their disposal, they pose a threat in a modern, open society. Even though the nuclear material available by theft or seizure might be far from optimal for a nuclear explosive, and a group would require shielded equipment to handle and process any intensely radioactive material, the threat still exists that plutonium, if available in sufficient quantities, could be used to make a crude device capable of at least a few kilotons of nuclear yield. While the yield would not be predictable for material containing much ^{240}Pu, the threat of spreading plutonium over a city by any explosive means would surely terrify the public. Of course, nations can take many measures to provide high security for key nuclear facilities and shipments, but such measures themselves could be unsettling to the public. As a rule of thumb, the fewer the shipments required, the less directly usable or convertible the material, and the less attractive the facilities as a possible terrorist target, the better.

For S&T systems, the terrorist scenarios present few features not raised in discussions during the evaluation by the International Nuclear Fuel Cycle Evaluation Group (1980). At that time, governments expressed confidence that terrorist attacks could be deterred or be resisted as necessary. Perhaps the biggest change is the more sober attitude of governments today about the possibilities open to determined terrorists and the difficulty of controlling them in an open democracy. One cannot do more than qualitatively assess the S&T proposals for this proliferation threat. Major fuel-processing facilities with large quantities of plutonium-bearing material in bulk form, which might be potential targets, may require isolated siting and high security like that of a military base. Also, the requirements for frequent fuel shipments would raise questions about transportation-related risks for most of the S&T proposals.

A safeguards study of an integrated ALMR/IFR with pyroprocessing suggests that such a facility is an unattractive target for a takeover and theft of material (Wymer et al., 1992). The material contained in the facility is less directly useful, and the sealed, self-contained site may make it easier to protect. Perhaps a similar integration of small, more readily guarded processing facilities at the reactor could be accomplished for S&T proposals that use aqueous technology. However, the kind of steps that make a facility minimally useful for converting material if the facility is seized, and enhance containment and surveillance techniques, tend to get in the way of reliable operation. In addition, it might be easier to protect a few large facilities than myriad small ones. For aqueous reprocessing, therefore, it is not clear that one would gain by going to multiple, small facilities.

Abrogation Threats

Abrogation raises mainly political issues. Technology may play a part in lengthening the time or reducing the extent to which the seized facilities may be used for weapon materials production. In the nightmare abrogation scenario, the nation trains its personnel at the facility in question, but secretly prepares elsewhere the rest of what it needs for a nuclear weapon, then seizes the facility, throws out the inspectors on some pretext, and prepares the bomb material it needs within a few days or weeks. Presumably, the more separated nuclear material a facility contains (i.e., more bombs could be produced) and the more readily the facility itself can be adapted to weapon materials production (i.e., the production time scale would be shorter), the more the perceived threat of abrogation would presumably increase.

Whether a nation would actually abrogate treaties and then misuse its civil nuclear resources has never been put to the test. The few nonnuclear-weapon nations credited with nuclear-weapon capability attained this capability using facilities that were outside a safeguards regime; other proliferation barriers did not deter them. However, an abrogation threat is very much a matter of perceptions.[9] Without full-scope safeguards, a nation might take over the facilities and obtain nuclear weapon materials without other nations being aware in time for effective counteraction. Even with full-scope safeguards, the presence of large reprocessing facilities and/or enrichment facilities in the S&T proposals tend to make them targets in national abrogation scenarios.

A new feature of the ALMR/IFR proposal is a reactor with integral onsite processing, where the nuclear material at the plant and the accessible plant equipment may not be well suited for rapidly obtaining weapon material. The IFR safeguards study indicates that a plant could be designed to reduce the risk under abrogation scenarios (see Appendix H). Unfortunately, the ALMR/IFR proposal has not yet been developed in enough detail to permit a definitive evaluation.

Conclusions

Nuclear proliferation is an issue even for the once-through fuel cycle, where it is addressed by domestic security measures and especially by international safeguards to deter the misuse of reactors, enrichment facilities, and stored spent fuel. Proliferation risks would generally be greater with widespread implementation of S&T systems in the many nations using nuclear power, mainly because of two factors: (1) the availability of bulk quantities of plutonium in separated or readily converted form at various places in the fuel cycle, which can be a challenge for safeguards even with stringent materials accountability and surveillance systems; and (2) the availability of large reprocessing facilities that could be misused for production of fissionable weapon materials, e.g., after treaty withdrawal or abrogation. The Clinton administration's policy announced in September 1993 reaffirms the link between U.S. nonproliferation goals and concerns vis-à-vis civil plutonium reprocessing and its use in nuclear power, as emphasized by the United States in the late 1970s, and discourages any S&T undertaking with LWR spent fuel in the United States for the foreseeable future.

HEALTH AND SAFETY ISSUES

Health Impacts

The health effects on the population as a result of exposure to radiation can be calculated. The analyst first defines the principal pathways by which radiation exposure could occur and then calculates just how much of the radioactivity can move through each pathway. Finally, it must be determined just how large a dose of radiation the exposed person would receive and what health effects it would produce. In most cases, the data required to perform such a calculation are not completely known, so there is considerable uncertainty in the final result of the calculation.

The collective population dose is the sum of the individual doses to a particular population. Doses of this type are expressed in units of person-sievert (1 pr-Sv = 100 pr-rem). Risk coefficients for various health effects have been developed over the last 50 years. For example, the BEIR V report (National Research Council, 1990) suggests that a collective dose-risk coefficient of 0.08 latent cancer fatalities/pr-Sv (8×10^{-4} cancers/pr-rem) can be postulated for a demographically average group following an acute dose. The report notes that accumulation of the same dose over weeks or months is expected to reduce the lifetime risk by a factor of 2 or more. This would give about 4×10^{-4} cancers/person-rem for the collective dose-risk coefficient for the case considered here.

Health Effects of Fuel Cycles

Various studies in recent years have quantified the health effects of various nuclear fuel cycles. Tables 6-2, 6-3, and 6-4 show the results of two of these studies, one done by the National Council on Radiation Protection and Measurements (NCRP) and the other by Oak Ridge National Laboratory (ORNL). The results of the NCRP study are expressed in terms of pr-Sv, however, and those of the ORNL study as the number of fatal health effects. By far the most likely fatal effects are latent cancer fatalities. (The columns labeled "general public mortalities [radiological]" are, for all practical purposes, latent cancer fatalities.) Table 6-5 puts these three study results on a common basis by converting pr-Sv to health effects. It also equalizes the different dose commitments; the NCRP calculation assumed a 100-year dose commitment where ORNL used 1,000 years.

It is interesting to note that the societal impact of any one of these fuel cycles is less than one latent cancer fatality per reactor-year. Note also that the generation of 20% of nuclear electricity by ALMRs would reduce mining and milling risks, since less uranium would need to be mined. On the other hand, since ALMRs recycle the actinides, they must use reprocessing, which introduces a risk not present

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[9] Several observers have speculated that the prospect of a future Iraqi abrogation of the NPT, with immediate seizure and reprocessing of accumulated spent fuel from the Osirak reactor, was a significant factor in motivating the 1981 Israeli air attack on the reactor before it went critical.

in the once-through cycle. The magnitude of these latent cancer risks is interesting, but the reader should be aware that the calculated risks vary depending on assumptions and have quite large uncertainties—perhaps a factor of 2 or more. The values from the two groups (shown in Table 6-5) are consistent with that level of uncertainty. Even with this level of uncertainty, it is evident that the difference in risk for the two fuel cycles is small.

TABLE 6-2 Summary of Collective Effective Dose Equivalents to the Regional Populations Normalized to a 1-GWe Reactor Operating 80% of the Time

Facility	Collective Effective Dose Equivalent (person-rem/yr)	Basis of Estimate
Mining	94	Weighted for two types of model mines (one-half open pit and three underground)
Milling	25	0.4 model mill
Conversion	0.03	Weighted for two plants, for 29.8 GWe
Enrichment	0.01	Paducah plus Oak Ridge for 29.8 GWe
Fabrication	0.004	Weighted for seven plants for 29.8 GWe
Reactor	4.8	1980 data for 47 plants
Low-level waste storage	—	No estimate available
Transportation		
(incident-free)	7.1	Excludes decommissioning wastes
(accidents)	5.4	
Total	136	

NOTE: 1 person-rem/yr = 0.01 person-Sv/yr.
SOURCE: National Council on Radiation Protection and Measurements (1987).

TABLE 6-3 Summary of Potential Health Risks Among the Total U.S. Population per GWe-year for the Once-Through LWR Fuel Cycle, Assuming 1,000-Year Dose Commitments

	Occupational Mortality.				Injury and Disease		
Source of Risk	Radiological	Nonradiological	General Public Mortalities (radiological)	Total Mortality	Occupational	General Public	Total Injury and Disease
Uranium mining	0.081	0.025	0.105	0.211	3.47	0.083	3.55
Uranium milling	0.012	<0.001	0.217	0.239	2.64	0.168	3.81
UF_6 conversion	0.001	<0.001	0.076	0.077	[a]	<0.001	0.001
Enrichment	0.002	<0.001	<0.001	-0.002	[a]	0.002	0.002
Fuel fabrication	0.054	<0.001	<0.001	0.054	[a]	0.06	—
Power generation	0.12	0.01	0.028	0.158	5.0	0.10	5.1
Transportation	0.001	0.01	0.061	0.072	0.17	[a]	0.17
Reprocessing	—	[a]	—	—	—	—	—
Waste management	[a]	[a]	0.008	0.008	[a]	0.016	0.016
Catastrophic accident	[a]	[a]	0.1	0.1	[a]	[a]	0.15
Total	0.271	0.045	0.595	0.921	11.28	0.429	11.8

[a] These values are not currently available, but they are expected to be small relative to those presented.
SOURCE: Michaels (1992; and private communication, 1993).

Health Effects of Transmutation

The three claimed beneficial effects of transmutation on radiological health risks are: (1) it lowers the incidence of harmful effects on health-related mining and milling activities by reducing the amount of natural uranium required to produce a given amount of electricity, (2) it lowers the long-term incidence of harmful effects on health of HLW disposal, and (3) it reduces the radiation exposure in some human intrusion scenarios. Offsetting these benefits to some degree is the added exposure from the recycling activity required for transmutation.

TABLE 6-4 Summary of Potential Health Risks Among the Total U.S. Population per GWe-year for the ALMR Fuel Cycle with Uranium Recycle, Assuming 1,000-Year Dose Commitments

	Occupational Mortality				Injury and Disease		
Source of Risk	Radiological	Nonradiological	General Public Mortalities (radiological)	Total Mortality	Occupational	General Public	Total Injury and Disease
Uranium mining	0.064	0.020	0.083	0.167	2.74	0.066	2.81
Uranium milling	0.099	<0.001	0.172	0.181	2.09	0.134	2.22
UF_6 conversion	0.001	<0.001	0.061	0.062	a	<0.001	0.001
Enrichment	0.002	<0.001	<0.001	0.002	a	0.002	0.002
Fuel fabrication	0.044	<0.001	<0.001	0.044	a	0.05	—
Power generation	0.12	0.01	0.028	0.158	5.0	0.10	5.1
Transportation	0.001	0.01	0.027	0.038	0.17	a	0.17
Reprocessing	0.002	a	0.068	0.109	0.01	0.12	0.13
Waste management	a	a	<0.001	<0.001	a	<0.001	<0.001
Catastrophic accident	a	a	0.1	0.1	a	a	0.15
Total	0.243	0.04	0.54	0.86	10.0	0.472	10.5

[a] These values are not currently available, but they are expected to be small relative to those presented.

NOTE: 20% of all nuclear electricity is assumed to be generated by ALMRs with recycle.

SOURCE: Michaels (1992, and private communication, 1993).

TABLE 6-5 Health Effects to Public for Fuel-Cycle Activities of LWR and ALMR per Gwe/year

Activity	NCRP 92[a]	ORNL[b] (LWR)	ORNL (ALMR)
Mining	0.21[c]	0.105	0.083
Milling	0.056[c]	0.217	0.172
Conversion	<0.001	0.076	0.061
Enrichment	<0.001	<0.001	<0.001
Fabrication	<0.001	<0.001	<0.001
Reactor	0.002	0.028	0.028
Transportation	0.005	0.061	0.027
Waste	na	0.008	<0.001
Accident	na	0.1	0.1
Reprocessing	0	0	0.068
Total	0.27	0.60	0.54

[a] The values from National Council on Radiation Protection and Measurements (1987) have been multiplied by a risk coefficient of 0.04 cancer fatalities pr-Sv (4×10^{-4} cancers/pr-rem)

[b] ORNL used a higher ^{235}U content in tails than National Council on Radiation Protection and Measurements, so more mining and milling is required per GWe.

[c] NCRP used a 100-year dose commitment, ORNL a 1,000-year commitment. The NCRP value has been multiplied by a 5.6 to obtain a 1,000-year value.

NOTE: The health effects of Tables 6-2 through 6-5 have been put on a consistent basis of expected latent cancers fatalities per GWe-yr of electricity.

The proposed introduction of ALMRs into the nuclear power mix of 20% would, in theory, reduce the number of latent cancers by 10%—from 0.6 to 0.54 per plant-year. This difference of 0.06 cancers/reactor-year converts to 1.5 pr-Sv per plant-year or 150 pr-rem per plant-year using a risk coefficient of 0.04 cancers/pr-Sv. According to NRC policy formulated in 1970 but still being followed, a reduction of public radiation dose is warranted if it can be obtained for an expenditure of $1,000/pr-rem or less or $150,000/plant/yr or less.

The cost for the reduction in dose to the population can be estimated as follows. In the economics section of this chapter, it was stated that the extra cost for using the ALMR amounted to about 8%. A 1,000-MWe plant operating 7,000 hours a year would produce 7.0×10^9K WH/yr. If the wholesale price of the electricity produced was $0.05/k WH, then the total value of the electricity would be $350 million. If the cost were 8% higher for the ALMR, the value of the electricity would be $350 million × 1.08 = $475 million or $28 million/yr more. The dose reduction noted above is 150 person-rem/yr. Using the NRC guidelines, the dose reduction change would be required if it could be obtained for $150,000/yr or less. Clearly, the $28 million/yr is far in excess of any amount the NRC would require. It is interesting to note that the annual background radiation dose to the U.S. population is about 0.3 rem/pr-yr × 250 × 10^6 pr = $75 million pr-rem/yr. This background dose is about 500 times greater than the population dose from the operation of 100 large nuclear power reactors.

Reduction of actinides turns out not to lower doses related to HLW disposal very much, since the LWR UO_2 fuel provides a good insoluble matrix for the actinides in a nonreducing environment. Transmutation of the actinides might lower the calculated dose for some human-intrusion scenarios. To justify the transmutation of actinides to reduce the human intrusion risk, there clearly would have to be a more detailed consideration of alternatives for decreasing the human intrusion risk. These options must include policy as well as technical solutions.

The reactor-based systems for reducing waste inventories by transmutation are specially fueled LWRs, fast reactors, and special-purpose reactors such as the PBRs. Fuel processing and fabrication facilities are required as well in support of these reactors. While much is known about the safety and environmental effects of LWRs, less is known when actinide fuels are involved. Fast-reactor safety is reasonably well understood, although experience with commercial operations is limited.

The only separations (fuel reprocessing) technology that is well established is PUREX.

Accelerator-driven waste transmuters have the advantage of subcritical operation, quick response times in the event of accelerator problems, and fission product inventory management (in some cases). Because they are still in an extremely early stage of development, very little is known about their environmental, health, and safety effects. Safety concerns relating to decay heat, inadvertent criticality, beam-target accidents, and system integrity have been raised.

The absence of a consistent set of risk assessments for the accelerator systems of waste management precludes quantitative comparisons with other transmutation concepts. However, it is possible to make some qualitative observations on comparative risk by considering the body of evidence reviewed in this report. That evidence generally supports the following very preliminary risk and safety rankings for the three categories of transmutation, separations, and disposal.

- For transmutation, the lowest-to highest-risk alternatives appear to be:

- —light-water reactors,
- —fast reactors, and
- —accelerators.

- For separations, the lowest-to highest-risk alternatives appear to be:

- —PUREX,
- —pyrometallurgical,
- —TRUEX, and
- —concepts involving on-line processing of liquid fuels.

- For disposal, the lowest-to highest-risk alternatives appear to be:

- —geologic repository and
- —surface.

The primary bases of the above qualitative rankings are risk analyses that have been performed on such operations as nuclear reactors, aqueous reprocessing, and, to some extent, geologic repositories. On the other hand, potential safety concerns have been identified—but little or no definitive risk assessment work has been performed—on accelerator-based systems, non-PUREX separation methods, and long-term surface storage facilities.

Transmutation and separation technologies have been proposed for the management of radioactive wastes on the theory that they have the potential to reduce the radiological risks of the once-through cycle by making more effective use of fissionable resources to generate energy. The studies referenced in this chapter do not show a large decrease in population dose because a fuel-cycle mix with 20% ALMR systems was used rather than the once-through cycle.

Summary

HLW separations and transmutation systems considered are: (1) reactor-based, (2) accelerator-based, and (3) the LWR once-through system. Each of these systems varies in the magnitude of the health and safety risk and the time period during which the public is most exposed. However, for the reactor-based cycles and the once-through cycle with good information bases, the total radiation exposure of the general public from the entire nuclear fuel cycle is very small. An ORNL report estimates that for the LWR once-through fuel cycle the mortality rate from fatal cancers among the general U.S. population, assuming a 1,000-year dose commitment, would be 0.6 mortality/GWe-yr, or 60 fatal cancers/yr for a 100 reactor industry (the size of the U.S. nuclear industry today). In a cycle where 20% of the electricity was generated by ALMR actinide burners, this rate was lowered to 0.54 mortality/GWe-yr. Using the same risk coefficients, one finds that natural background radiation would produce over 20,000 fatal cancers per year in the U.S. population. The risk levels in the various fuel cycles could be further reduced by improvements in mining and milling if needed. Thus, the radiation risk levels in the whole fuel cycle are very small for either waste management approach.

In the once-through cycle for long time periods of 10^6 to 10^8 years, ^{129}I and ^{99}Tc dominate the risk because of their high solubility compared to that of the actinides. However, in some intrusion scenarios, actinides do dominate the risk. In these cases, transmutation of the actinides would reduce the risk.

Information needed to make a meaningful estimate of the fuel-cycle risk for ATW is not available, so no comparison can be made at this time.

COST OF FUEL REPROCESSING

Introduction

A major contributor to the fuel-cycle cost of any of the proposed transmutation concepts is fuel reprocessing. Costs are incurred not only from reprocessing the LWR fuel to recover the actinides and other radionuclides to be transmuted, but also from the multiple reprocessing required to expose these LWR-recycled radionuclides to many irradiation cycles until they are sufficiently transmuted.

The transmutation proposals project commercial operation of the transmutation reactors during the period 2005 to 2012. The proposals predicate that many large power plants fueled by actinide recycle and the associated reprocessing facilities would be financed and constructed by private industry. To meet the proposed schedule of deployment, the electric utility industry and the chemical industry would have to commit to detailed design, financing, licensing, and construction of new actinide-fueled power plants and of associated reprocessing plants far in advance of the first commercial-scale operation. To furnish TRUs for starting a transmutation reactor, the new reprocessing plant for high-yield recovery of all actinides from spent LWR fuel would have to begin operation well in advance of the first commercial transmutation reactor.[10]

For example, DOE's ALMR program proposes a centralized commercial facility capable of reprocessing 2,700 Mg/yr of LWR spent fuel. It could supply the actinides for the annual introduction of one 1.4 GWe ALMR. Operation would have to begin about 2 years prior to the start-up of the first ALMR, assuming a year for reprocessing and a year for fuel fabrication. The ALMR program also estimates the cost of a plant to reprocess LWR spent fuel with capacity as low as 300 Mg/yr. This smaller plant would have to begin operation as early as 9 years in advance of the first commercial ALMR. Similar lead times would be required for other transmutation systems, such as LWRs and ATWs. Such early deployment of high-yield LWR fuel reprocessing would require well-established technology for these new facilities and operations. Industry must have a strong basis on which to commit risk financing, not only for the full-scale transmutation power plants but also for the new reprocessing facilities.

Of the various processes proposed, aqueous reprocessing has the best chance of being established in time for the ambitious schedules proposed for transmutation. The status and possible cost of conventional aqueous reprocessing with U.S. financing are reviewed here, utilizing information on costs for contemporary reprocessing plants in France, the United Kingdom, and Japan. How the reprocessing facilities are to be financed has a large effect on the unit cost of reprocessing. The issues discussed herein would also apply to pyrochemical separations, once the pyrochemical process design and plant design have proceeded sufficiently to yield a reliable basis for cost estimates.[11]

The greatest uncertainties of feasibility and cost of the ALMR system appear to be in reprocessing.[12] The ALMR program has estimated costs for high-yield recovery and recycle of TRUs from LWR spent fuel, both for Argonne National Laboratory's (ANL's) proposed pyrochemical reprocessing and for conventional aqueous reprocessing. However, there are enormous disparities in the estimates made by transmutation projects, even in the cost of conventional aqueous reprocessing, assuming new commercial-scale reprocessing plants are to be constructed in the United States.

In this chapter, the costs for contemporary aqueous reprocessing plants in France, the United Kingdom, and Japan are reviewed, and the data are used to estimate the cost of conventional aqueous reprocessing if constructed now in the United States. The effects of various methods of financing the reprocessing plant (i.e., financed by government, electric utilities, or private industry) are shown. These estimates are then used as benchmarks to compare with reprocessing

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[10] Transmutation/reprocessing proposals from DOE laboratories specify high-yield recovery and recycle of all TRUs to achieve the claimed benefits to waste disposal. As pointed out earlier in this chapter in the section *Impact of S&T on Waste Repository*, waste disposal in some repository settings could be benefited even by lower-yield recoveries of plutonium without recycle of MAs if some cost-effective means to reprocess LWR spent fuel were available. The reference cost analyses adopted for this study are based on such conventional reprocessing technology.

[11] The Argonne National Laboratory prefers pyroprocessing technology both for spent LWR fuel and for ALMR spent fuel. However, General Electric's estimates of fuel-cycle costs for the ALMR program are apparently based on the cost of aqueous technology for reprocessing spent LWR fuel.

[12] If one were to adopt the more optimistic costs of LWR fuel reprocessing, such as a recent estimate of $165/kg quoted by a member of a STATS subcommittee (M. Coops, private communication, 1994), the main contribution to overall transmutation costs would be from capital costs of the transmutation reactor, and small uncertainties in reprocessing costs would not be as important. However, if the costs of reprocessing LWR spent fuel to obtain TRUs even for a breeding ALMR were as high as estimated herein for commercial aqueous reprocessing in the United States, the very concept of operating an ALMR to be started and fueled with TRUs from LWR spent fuel would be economically untenable. As shown later, ANL estimates (Chang, 1993) that if the cost of LWR reprocessing were greater than $350/kg, it would be cheaper to start and fuel the ALMR on enriched uranium rather than on the TRUs in LWR fuel recovered for the purpose of transmutation assuming no credit is taken for reductions in risk or cost of disposal. The reference reprocessing cost estimated herein for the private financing proposed by DOE would be 600% too great.

cost estimates made by the transmutation projects.

A more detailed analysis of the cost of reprocessing, together with historical perspective of commercial fuel reprocessing in the United States is presented in Appendix J.

Relative Economics of Reprocessing versus Once-Through Fuel Cycle for LWRs

Since all of the transmutation concepts would involve reprocessing and recycle of actinides from LWR spent fuel, it is instructive to examine the cost of conventional aqueous reprocessing to recycle uranium and plutonium in the LWR fuel cycle, even neglecting the expected higher cost of high-yield recovery and recycle of all the actinides as proposed by the transmutation concepts.

A recent study of LWR fuel-cycle costs by the Nuclear Energy Agency (NEA) of the Organization of Economic Cooperation and Development (OECD) (OECD/NEA, 1993) concluded that the levelized fuel cost for the once-through LWR fuel cycle is approximately 14% less than for the reprocessing cycle. The OECD/NEA study assumed low-risk government financing. The once-through non-reprocessing fuel cycle would be even more economically favorable in the United States if, as proposed by U.S. proponents of transmutation, the reprocessing facilities were owned and financed by private industry.

Those countries that have chosen to reprocess their nuclear fuel have evidently based their decision on a number of factors, in addition to cost. With limited natural uranium resources, for example, they have a strong interest in being energy self-sufficient. Also, some countries expect to take an early write-off on the large capital cost of their new reprocessing plants, so that sunk costs no longer contribute to the cost of further reprocessing.

Capital Costs and Throughouts of Contemporary Reprocessing Plants

Capital cost information was obtained from the open literature and from private communications, based on data reported for three reprocessing plant projects: THORP (U.K.), UP3 (France), and Rokkashomura (Japan). UP3 and THORP are in operation, and Rokkashomura is under construction. According to data reviewed in Appendix J, the annual throughputs of all three plants are in the range of 800 to 900 Mg/yr, about threefold less than the 2,700-Mg/yr throughput proposed by the ALMR project.[13] All provide complete services ranging from fuel storage through HLW vitrification. Based on the detailed data reviewed in Appendix J, the costs in U.S. dollars for constructing plants of 800 to 900 Mg/yr throughput in the United States, not including interest during construction, are shown in Table 6-6.

TABLE 6-6 Estimated Capital Costs a for Contemporary Reprocessing Plants

Primary Source of Data	THORP (UK)	UP3-La Hague (France)	Rokkashomura (Japan)
Annual throughput, Mg/yr	900	800	800
Capital cost, $million (in 1992 $)[a]	5,370	6,670	6,500

[a] Financing costs, including interest during construction, are not included. See Appendix J for details and references.

The capital cost for the 900-Mg/yr plant was derived from the 1993 study by OECD/NEA, *The Economics of the Nuclear Fuel Cycle*, with details shown in Table 6-7. The OECD/NEA study is based on cost data from the recently completed THORP plant and includes input from COGEMA, the owner-operator of UP3, the most recent French reprocessing plant. The total estimated cost of £3,297 million ($5,370 million) is 15% higher than the reported actual cost of THORP of £2,850 million ($4,560 million) without interest during construction. The reason for this cost difference is not evident, but it could be associated

TABLE 6-7 OECD/NEA Reprocessing Plant Capital Cost Estimate (900 MTHM/year annual throughput)

Cost Component	Capital Cost (1991 £ millions)
Fuel receipt and storage	100
Reprocessing plant	2,300
High-Level waste	
Vitrification	260
Interim storage	59
Intermediate-level waste	
Encapsulation	300
Interim storage	38
Site preparation and services	
Site preparation	229
Site services	11
Total capital cost	
1991 £ (million)	3,297
1992 $ (million)	5,370

NOTE: Does not include interest during construction.

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[13] As explained in Appendix J, the expected annual throughout is lower than the design (nameplate) capacity, due to downtime typical for commercial fuel reprocessing. However, there are apparently some individuals who expect less downtime and higher throughput (A.G. Croff, private communication, 1994).

with the higher cost of construction at a "grass roots" site instead of at the existing site of THORP.[14] , [15]

Operating Costs

The following analysis of operating costs of an aqueous reprocessing plant uses the OECD/NEA operating cost estimate of £230 million per year, based on THORP data and already subjected to peer review [OECD/NEA, 1993]. This includes vitrification and interim storage of HLW; encapsulation, interim storage, and disposal of intermediate-level waste; and disposal of LLW. Applying the 10-year average currency conversion rate (1983-1992) results in an annual operating cost of $374 million for 900-Mg/year throughput.

Financing Costs

Estimating unit costs of reprocessing from the capital and operating costs developed above requires definition of the financial structure of the prospective plant owner/operators and the associated costs of debt and equity capital. The various types of organizational entities considered here include: private venture, industry-utility consortium, and government. Each of these arrangements would involve a different cost of financing, reflecting in part the risk of realizing an adequate return on investment.

Private Financing

DOE assumes that private industry would finance, construct, and operate a large (2,700 Mg/yr of LWR spent fuel) centralized reprocessing plant in order to obtain TRUs from spent fuel to start and refuel TRU-burning ALMRs. The plant would have to begin operation in 2010 to furnish TRUs to start the first large commercial ALMR proposed for introduction in 2012. However, a purely private venture to design, build, own, and operate such a complex without government financial guarantees, as might be undertaken by the chemical industry, appears unrealistic. The level of financial risk appears unacceptably high to obtain the required private financing in the United States due to

- immature, unproven technology;
- likely strong opposition from public-interest groups;
- regulatory uncertainty (first-of-a-kind facilities);
- general reluctance of the financial community to finance nuclear projects;
- Potential adverse changes in government policy that could preclude plant operation; and
- experience with the Barnwell reprocessing plant, a major deterrent to new ventures in private reprocessing.

The new reprocessing plant would be based on presently unproven pyrochemical technology, operating with over tenfold lower process losses than yet achieved in commercial reprocessing facilities. To justify the market for the TRUs recovered from this new reprocessing plant, the utilities would have to commit to build over 20 large ALMRs. There would be no other market for the TRU product from the reprocessing plant. Being highly contaminated with fission products and minor actinides (neptunium, americium, and curium), the TRUs could not be used as MOX fuel in current LWRs.

Even discounting the unusual risks if private industry were to finance such a reprocessing venture, private financing would require a much higher return on investment. As shown in Table 6-8, an annual fixed-charge rate of at least 20% per year would be required, almost twice that for utility financing and over threefold greater than that for government financing. The fixed-charge rates shown in Table 6-8 are based on an inflation-free economy.

Utility Financing

The rationale for utility ownership of a project requiring both a special-purpose, non-LWR reactor technology to transmute radionuclides *and* an integrated fuel reprocessing plant based on first-of-a-kind, nonproven technology is difficult to understand.

The Nuclear Waste Policy Act of 1982 essentially relieves utilities of the responsibility for postirradiation processing and permanent disposal of spent fuel. Utilities currently have little incentive to become involved as an owner in a high-risk reprocessing venture.

As shown in Table 6-8, the annual fixed-charge rate for utility financing would be about 12.3% per year, based on an inflation-free economy.

Government-Owned Reprocessing Facility

Ownership of reprocessing plants in European countries has generally been confined to government corporations. They possess the unique ability to finance projects at low interest rates and are able to undertake complex, high-risk

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[14] NEA lists a number of factors that could reduce the capital cost. However, additional costs would incur if the reprocessing facility described in the OECD/NEA study were to be modified for high-yield recovery of all actinides and if it were later extended to the recovery and recycle of TRUs from multiple recycled fuel.

[15] The OECD/NEA cost estimate relies on data for the small number of commercial reprocessing plants that have been constructed, mainly in the United Kingdom and France. For a transmutation program that must span many decades and centuries to accomplish the stated goals, a large number of such plants would have to be constructed, including several later generations of plants to replace those that will have reached their design life. Some eventual saving from standardization would be expected.

projects in the interest of national policy. Without such low-cost financing from the government or from advance payments for services by reprocessing customers, as was the case for the U.K.'s THORP plant, reprocessing costs would be quite high. Since the U.S. federal government is ultimately responsible for the safe disposal of spent fuel in the United States, it would be the logical entity to undertake a TRU-burning program, if this were justified and possible within the institutional barriers of government.

TABLE 6-8 Reference Case Parameters

Parameters	Government	Utility	Private Venture
Cost Assumptions			
Capital cost ($ M)[a]	6,160	6,670	7,320
Operating Cost ($ M/yr)	374	374	374
Property taxes and insurance (%/yr of initial capital cost)	0	2	2
Annual refurbishment cost (%/yr of initial capital cost)	1	1	1
Financial Structure			
Common stock (%)	0	46	70
Preferred stock (%)	0	8	0
Debt (%)	100	46	30
Investment Returns and Interest Rates (constant $)			
Common stock (%/yr)	na	8.5	16.0
Preferred stock (%/yr)	na	4.1	na
Debt (%/yr)	4.0	4.8	9.0
Weighted cost of capital (%/yr)	4.0	6.4	13.9
Tax Assumptions			
Income tax rate (%)	na	38	38
Tax recovery period (years)	na	15	15
Investment tax credit (%)	na	0	0
Book life (years)	na	30	30
Annual Fixed Charge Rate (%)[b]	5.8	12.3	20.8

[a] Includes interest during construction.

[b] The annual fixed charge rate is the percentage of the total capital investment that is charged yearly against the cost of electrical energy produced, based on an inflation-free economy. It is derived from date on financial structure, investment returns and interest, and tax assumptions. (See reference EPRI (1993) in Appendix J).

As shown in Table 6-4, government financing could require an annual fixed-charge rate as low as 5.8% per year, based on an inflation-free economy.

Unit Costs of LWR Reprocessing with U.S. Financing

For this study unit reprocessing costs were calculated for a conventional aqueous reprocessing plant with an annual throughout of 900 Mg/yr, if owned by the government, utilities, or by private industry. An overall capital cost of $5,370 million was adopted, based on the new THORP facility, as shown in Table 6-7. To calculate interest during construction, a total project schedule of 9 years was assumed. The estimated annual operating cost is $374 million. The calculated unit costs are given in Table 6-9. Thus, U.S. private financing would result in a unit reprocessing cost over twice that for government financing.

The unit cost with U.S. private financing would be even greater if the capital costs for France's UP3 and Japan's Rokkashomura are adopted. Both plants have an 800-Mg/yr throughput. Operating costs are assumed to be proportional to throughput. The unit costs with U.S. private financing are estimated to be $2,860/kg for UP3 and $2,640/kg for Rokkashomura. There is a variation of about 36% in unit reprocessing costs estimated for the three contemporary plants, if constructed with U.S. private financing. However, as shown in Table 6-9, the uncertainty in reprocessing costs associated with ownership and financing is much greater than the variation among the contemporary plants.

The unit costs of reprocessing are all considerably greater than the calculated break-even costs for reprocessing LWR spent fuel. For example, ANL (Chang, 1993) calculates that if the unit cost of reprocessing LWR spent is greater than $350/kg, it would be cheaper to start and fuel ALMRs on enriched uranium rather than on TRUs recovered from reprocessing LWR spent fuel.[16] Thus, if the cost of reprocessing LWR spent fuel were to exceed $350/kg, and if the utility owner were to make decisions on the basis of fuel-cycle costs without subsidy, the more economical fuel cycle based on enriched-uranium start-up would be chosen. Consequently, without federal subsidy to the utility, the fuel cycle involving reprocessing LWR spent fuel to transmute actinides in commercial ALMRs would not be chosen, and the claimed benefits to geological disposal would not be realized. If, hypothetically, actinide burning in ALMR were to yield a large economic benefit to waste disposal, this could be the basis for a federal subsidy that could turn the decision towards fueling the ALMR with actinides from LWR spent fuel. However, the cost analysis presented herein suggests that the claimed economic benefits would not be likely if the real reprocessing costs were to be in the neighborhood of the costs calculated for conventional commercial reprocessing.

TABLE 6-9 Calculated Unit Cost for Conventional Aqueous Reprocessing, 900 Mg/yr, U.S. Financing

Plant Owner/Operator	Unit Cost of Reprocessing, $/kg
Government	800
Utility	1,300
Private industry	2,100

Costs Associated With Other Reprocessing Plant Studies

A number of other studies of the cost of aqueous reprocessing plants have been reported.

- The 1990 study for a generic U.S. site (Gingold et al., 1991) estimated reprocessing-plant capital costs ranging from $2,725 million (government owned) to $3,001 million (privately owned) for an annual throughput of 1,500 Mg, over 60% larger than the 900-Mg/yr throughput of THORP. These costs assumed a mature industry.
- The ALMR fuel-cycle assessment (Taylor et al., 1992) developed a capital cost of $6,100 million for a 2,700-Mg/yr reprocessing plant, over threefold greater than the capacity of THORP. This cost also includes facilities for MOX fuel fabrication.
- An OECD/NEA Expert Group (OECD/NEA, 1993) derived unit reprocessing costs based on THORP data for plants of 900 Mg/yr capacity. The corresponding capital cost would be in the range of $6,000 to $7,000 million.

The capital costs estimated by Gingold and GE (Gingold et al., 1991; Taylor et al., 1992) are substantially below those experienced in the construction and operation of actual plants, even though the throughputs in those studies were two to three times higher than for THORP, UP3, and Rokkashomura. Assuming that capital costs are proportional to the 0.6th power of the plant capacity, Gingold's estimates scaled to 900 Mg/yr would result in a capital cost of $2,010 million for a government-owned facility and $2,210 for a privately owned facility. Similarly, the GE capital cost would decrease to $3,160 for a 900-Mg/yr plant, from which must be subtracted the cost of the fuel fabrication facilities that were included in the GE estimate. The costs estimated by Gingold and GE, adjusted to 900-Mg/yr throughput, are only a third to a half of the costs reported for actual contemporary plants of 800 to 900-Mg/yr capacity.

Even lower estimates of capital costs were presented earlier by the ALMR project (Salerno et al., 1989) for PUREX/TRUEX reprocessing plants designed for high-recovery yield of all TRUs from LWR spent fuel. Escalating their estimated from 1989 to 1992 for 300-Mg/yr throughput and $4,250 million for 2,500-Mg/yr throughout. Scaling to the cost for a reference plant with 900-Mg/yr throughput as above, the estimated capital costs would be $440 million derived from GE's 300-Mg/yr estimate and $2,200 million derived from GE's 2,700-Mg/yr estimate. If these estimates were reasonable, the smaller 300-Mg/yr plant would yield the lowest unit cost when compared to other estimates of contemporary costs of aqueous reprocessing reported in the GE study. Such an inverse economy of throughput has not been observed or predicted in other studies.

Based on the above information, it can be concluded that the reported capital costs for actual contemporary plants provide the most reliable basis at this time for estimating the cost of future plants. Estimated capital costs reported in recent U.S. studies are, in our judgement, optimistically low.

The Exxon commercial reprocessing plant, designed over 15 years ago by Bechtel but not yet built, is the last U.S. commercial reprocessing plant designed to sufficient detail to justify reliable cost data for that era. The U.S. experience and capability for design and reliable cost estimating of industrial-scale fuel reprocessing plants was then largely vested in the DuPont company and in Bechtel. DuPont designed, built, and operated defense reprocessing plants at Hanford and Savannah River, and Bechtel designed and built the commercial plant at Barnwell and designed the Exxon plant. Now, over a decade later, with little or no U.S. activity in engineering design of industrial-scale fuel reprocessing plants, the U.S. capability has eroded. We must look to recent foreign experience for guidance on costs.

In a 1983 paper, Wolfe and Judson (1983) noted that the

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[16] This assumes availability of highly enriched uranium (HEU) that is not being produced now and may not be produced in the future.

estimates of LWR unit reprocessing costs in constant dollars increased substantially from 1975 through 1983, as shown in Figure 6-2. Also shown in Figure 6-2 are the current estimates of the unit costs for reprocessing plants constructed in the United States, derived from estimated costs for contemporary plants in the United Kingdom, France, and Japan. Financial parameters were applied for a private venture in the United States, assuming optimistically that the financing would be characteristic of a low-risk project in the chemical industry. The unit costs are expressed for an inflation-free economy. The unit costs estimated for these three sources fall on extensions of the band of reprocessing costs shown by Wolfe and Judson, but at a level several-fold higher. It is clear that what may be financially valid for a government-owned European plant financed with customer prepayments for reprocessing services, and with relatively low annual charges on capital investment, is not necessarily applicable to the same plant constructed by private industry in the United States.

Also shown in Figure 6-2 are recent estimates by the ALMR project of unit costs for privately reprocessing LWR fuel financed by U.S. construction (Taylor et al., 1992; Chang, 1993). Each plant has a throughput of 2,700 Mg/yr, with high-yield recovery of all actinides and volatile fission products. The estimated unit costs are about $500/kg for aqueous reprocessing and $350/kg for pyrochemical reprocessing. The latter is about six-to eightfold below the estimated unit cost of a 800- to 900-Mg/yr U.S. plant, based on contemporary plant costs in the United Kingdom, France, and Japan.

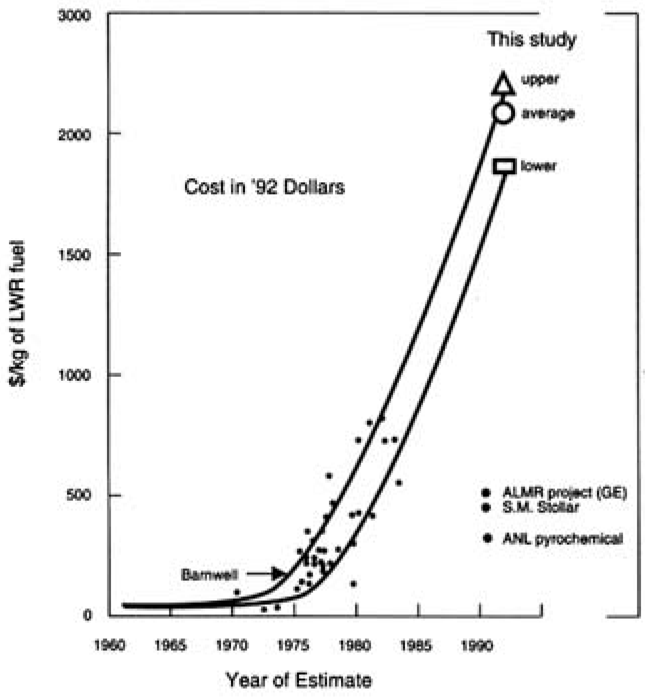

FIGURE 6-2 Current estimates of the unit costs for reprocessing plants constructed in the United States.

Also shown in Figure 6-2 are the unit costs estimated in 1991 by S.M. Stoller Co. (Gingold et al., 1991) for aqueous reprocessing and pyrochemical reprocessing, with U.S. private financing. The Stoller estimates, in a study financed by the Electric Power Research Institute (EPRI), are close to those made by GE, but far below the estimates derived herein from the new United Kingdom, French, and Japanese plants.

Comparisons with published reprocessing prices and with other estimates of reprocessing costs are given in Appendix J.

Summary

Costs of contemporary aqueous reprocessing plants in the United Kingdom, France, and Japan are important benchmarks to compare with U.S. estimates of reprocessing. For the purpose of this report, we adopt the OECD/NEA estimates of the capital and operating costs of a plant with 900-Mg/yr throughput. These are based largely on the U.K.'s THORP data, with input from France's COGEMA. We have translated these costs to U.S. construction as described in Appendix J: 10-28 and J: 37-60. We estimate interest during construction and calculate the unit reprocessing costs for a similar U.S. reprocessing plant for three forms of U.S. financing: government, $810/kg; utility, $1,330/kg; and unregulated private industry, $2,110/kg. Each of these costs is so high that there would be no financial incentive for operating a transmutation system that would require reprocessing spent fuel from LWRs, unless it were subsidized by the government for possible benefits to waste disposal. To obtain the high recoveries to recycle all the TRUs (not only Pu), as proposed by DOE laboratories and contractors, the cost of aqueous reprocessing would be even greater. Even higher costs for a U.S. reprocessing plant would occur if the delays experienced by previous reprocessing ventures were again encountered.

We have compared these unit costs derived from contemporary plant data with costs of aqueous reprocessing projected in studies by DOE laboratories and contractors for the purpose of transmutation economics. The latter are so much lower than those estimated in the present study that there is good reason to question the validity of all the recent U.S. estimates for the cost of reprocessing LWR spent fuel. Given that those estimates for aqueous plants are so far below the costs inferred from the European and Japanese benchmarks, it is questionable that reliable estimates could now be made of the pyrochemical process for LWR spent fuel, which is in a relatively early stage of development.

POLICY, INSTITUTIONAL, AND MANAGEMENT ISSUES

When it was first instituted, the Atomic Energy Commission (AEC) had both developmental and licensing authority over civilian nuclear facilities. Recognition of the embodied conflict of interest and increasing public concern over nuclear issues caused the splitting of the AEC into an energy research and development agency and a separate regulatory agency (the U.S. Nuclear Regulatory Commission), which led to greater public involvement in the licensing process. One result was an extended period during which requirements placed on nuclear facilities were changing even as the facilities were being built. These changes greatly increased costs and prolonged construction schedules for many nuclear facilities, and in some cases, provoked outright cancellation. Requirements were changed at the regulatory level (e.g., retrofits to reactor safety systems and the West Valley Fuel Reprocessing Plant) as well as at the policy level (e.g., President Carter's decision to oppose reprocessing).

Licensing and construction of major new nuclear facilities is at a standstill in the United States. This results in part from an adequate immediate supply of electrical generating capacity and a current surplus of uranium (resulting in a lack of need for reprocessing and recycling facilities). However, a factor at least as important is the reluctance of investors (utility or industrial) to risk large sums of money to build nuclear facilities because their investments might be jeopardized through regulatory or policy changes. This concern appears to be so widespread that it is unlikely any investor will attempt to build a major nuclear facility without guarantees from the federal government amounting to the project being government financed. This outlook can be expected to persist until a sufficient number of nuclear facilities have been built over a sustained period of regulatory stability to restore investor confidence.

Policy

Long-term major national commitments with strong, stable, centralized government leadership and financing and operating within a favorable public perception of nuclear enterprises will be required if S&T is to become a significant factor in nuclear waste management. On the basis of the history of the past decade of nuclear activities, it is difficult to visualize any such circumstances in the immediate future. Given this reality, it is necessary to consider (1) under what conditions future policymakers would find it prudent to undertake to implement S&T and (2) whether those conditions are likely enough to occur that research should continue, and if so, at what level.

The following are some of the conditions that might lead policymakers to advocate S&T and to support the legal, financial, and managerial provisions needed to bring it to fruition:

- if regulators or the public considered the technical estimates of repository risks to be too uncertain or too high, and if the estimated risks or uncertainties could be reduced to acceptable levels by improving the waste form or by changing or reducing the radioactive constituents of the waste;
- if difficulty in building the first repository made building a second repository undesirable or infeasible, and the limitation on amount of spent fuel that could be placed in the first repository could not be altered;
- if the federal government were unable to build and license a repository for institutional or technical reasons;
- if economic, fuel-supply, and societal conditions changed, so that it was decided to reprocess to recover fissile material, presenting the opportunity to transmute some constituents as a waste-management measure;
- if research uncovered an S&T system that did not present the problems inherent in those currently proposed; or
- if attitudes about nuclear power changed, perhaps as a result of concern about the worldwide environmental effects of other energy sources.

None of these conditions exists at present. However, one or more might come into being during the long period of time this committee estimates will ensue before economic and energy-supply conditions make S&T a practical economic alternative to the once-through reactor fuel cycle. In anticipation of such an eventuality occurring, a carefully planned but focused research program on S&T should be carried on.

Licensing, Siting, and Public Acceptance

Creation of a S&T system would require legislative and administrative steps establishing the policy basis for a national commitment to large expenditures for the many facilities needed. The operations of the various facilities would link numerous organizations over a period of many decades. Siting restrictions imposed by certain state governments would have to be relaxed. Creating and reviewing an environmental impact statement for an S&T system may be a more contentious undertaking than was the process for drawing up the Generic Environmental Statement on Mixed Oxides, which was discontinued in the mid-1970s.

Difficulties such as the lack of demonstrated need, economics, technical uncertainties, and particularly those associated with licensing and siting, are becoming increasingly important impediments to concrete actions in nuclear matters. Mechanisms are available through which opposition can influence policy and action more effectively than before: state laws and regulations in a domain that was historically federal; controls over nuclear matters at the state level through controls on nonnuclear hazardous materials criteria such as the Resource Conservation and Recovery Act (RCRA); and a continuing series of intervention lawsuits. Finally, the public's distrust of the ability of the federal government to conduct programs effectively undermines its capacity to organize, manage, and finance a program as large as would be required for S&T implementation.

There are many recent examples of the effects of the difficulty in completing nuclear-related projects:

1. A decade will be needed for the licensing and certification of Advanced Light-Water Reactors, even though they are a direct advance from 100 presently operating LWRs in the United States and have vastly improved safety characteristics.
2. The government has invested over $1.6 billion through April 1995 in Waste Isolation Pilot Plant (WIPP), a deep underground salt bed repository in New Mexico that was started in January 1975 and is now over 20 years old. The repository facility was fully constructed and capable of receiving waste in October 1988. After extensive review, DOE declared it ready for safe operation and authorized it to receive several small amounts of waste for bin tests deposits in about 1991. However, this operation was never started. It is still not operational as of the time of this report.
3. The continuing delays in the planned operation of Yucca Mountain are indicative of the difficulties licensing and siting a complete S&T system could encounter.
4. No LLW disposal site proposed since the Low-Level Waste Policy Act of 1980, amended in 1985, is operational. All proposed sites under the state compact agreements have been challenged and delayed.
5. Although DOE was expected to take custody of spent fuel from commercial reactors in 1998, it is widely assumed that this schedule will not be met. Utilities are working now to find a way to store fuel on site indefinitely.

Thus, in the present context of institutional relationships in the United States, developing, siting, and licensing an entire system of high-technology nuclear facilities, for which few precedents exist, are likely to take a very long time, cost a great deal, and continue to lack the assurance of eventual success. Any decision to build and operate an S&T system must take these constraints into account.

Since no systematic effort has been undertaken to gauge public opinion on transmutation, McCabe and Colglazier (1992) concluded, in a report for the committee's introductory workshop, that any characterization of public opinion on the subject is largely speculative. Newspaper and magazine articles on the subject of S&T have largely been based on press releases and interviews with proponents. These articles generally describe these proposals favorably, as potential technical solutions to the disposal of radioactive waste. The articles contain little or no discussion of the technical uncertainties, the potential management, financial, and institutional complexities, and the length of time needed for the research and operations that this committee has identified as barriers to implementation of current proposals. Thus, any current public support that may exist for S&T is likely to be based on very incomplete information. Before S&T proposals are implemented, the public will learn more about the great size, complexity, and number of nuclear-based facilities necessary to implement any of the current S&T proposals.

Financial Provisions

The facilities listed above—reprocessing, fuel fabrication, and transmutation—will be expensive to build and operate, as recent experience with even the simplest nuclear plant demonstrates. The first cost of any one of them is likely to be several billions of dollars. Moreover, the operation of an S&T cycle is certain to be more expensive than the once-through fuel cycle, until the price of uranium fuel makes the actinides from LWR fuel an economical substitute fuel source (which is many decades in the future). Thus, financing and supporting the entire system is not likely to be financially practical for any one utility, or any industrial group, without government subsidies or guarantees. In the end, any extra costs beyond those of the current fuel management system must be borne by electricity users or taxpayers. The fact that these incremental costs will have to be incurred in the next two to three decades to reduce the potential risk to society for hundreds to hundreds of thousands of years in the future, while short-term health risks may also be increased, will make the large-scale, long-term financing support needed for an S&T system difficult to obtain and ensure.

An estimate can be made of the time scale and the cost required to deploy an S&T system based on LWRs or ALMRs, assuming that development can be completed and that favorable institutional arrangements can be achieved. The licensing, construction, and initial operation of a S&T system of sufficient scale to ever begin to affect spent fuel emplacement in a geologic repository would require one to two decades after a system feasibility demonstration, and an expenditure of $20 to $40 billion beyond the costs of development and demonstration. Additional time and a much larger investment of funds would be necessary to start up an S&T system of sufficient scale to reduce repository hazards significantly or to affect the need for a second repository.

If electric utilities, chemical companies, and other industrial organizations are to participate as they have in the past deployment of nuclear power reactors and fuel-cycle facilities, many private organizations and their state and federal regulatory bodies must coordinate their plans, decisions, and expenditures to ensure the continuing integrity of the S&T system. Indeed, private utility funding presumably would be contingent on agreement by the many state public utility commissions that such long-term commitments are prudent investments. In view of the history of nuclear power and commercial reprocessing in the United States, federal guarantees are likely to be necessary for such industries to be involved with the construction and operation of the required S&T facilities.

System Requirements

The S&T system required to achieve the prospective benefits would comprise many interdependent components, e.g., waste-transmutation reactors, spent-fuel reprocessing plants, recycled fuel fabrication plants, plants to package the residual waste for ultimate disposal, and mechanisms to transport fuel and waste between the facilities. Developing, building, and operating selected individual components of the system would yield little benefit. A complete system incorporating many facilities, all successfully operating in a synchronized fashion for many decades to centuries, would be required in order to have a significant effect. If any one of the components of the system cannot be developed, licensed, and operated with reasonably high reliability, the desired benefits will be foreclosed.

Implementation of S&T technology would require the construction and licensing of the following four new types of fuel-cycle facilities in addition to those currently used or planned for a once-through LWR fuel cycle:

- LWR fuel reprocessing: One or more large plants would be required to receive LWR spent fuel, reprocess it to recover the radionuclides to be transmuted, and treat wastes in preparation for transportation and disposal. These might be based on existing technology (e.g., aqueous fuel reprocessing), pyrochemical reprocessing, or other technologies to be determined.
- Transmutation devices: A number of devices would be needed to transmute the recovered radionuclides. These might be LWRs at the outset, succeeded or augmented by one or more advanced transmutation devices such as those discussed in Chapter 4. To a large extent, these would substitute for other electricity-generating plants.
- Actinide/fission-product fuel/target fabrication: S&T involves transmutation of the TRU and fission-product radionuclides recovered during reprocessing. Transmutation requires that these radionuclides be fabricated remotely into an acceptable fuel or target for insertion into the reactor. In some cases, such as the fluid-fueled ATW, the fuel would always remain in slurry or solution form. In most other cases, the radionuclides must be fabricated into metal-clad rods containing oxides or metal.
- Transmutation-device fuel/target reprocessing: After irradiation, the transmutation-device fuel or targets must be reprocessed to recover the untransmuted radionuclides for refabrication and continued irradiation and to send undesirable transmutation products to the waste stream. This facility might be similar to what is used for reprocessing LWR fuel or it might employ an entirely different technology. For most systems, the reprocessing facility is physically disconnected from the reactor (and usually geographically separated), but in the case of the ATW, the reprocessing plant is integrated into the reactor design.

In any of the S&T systems the time required to make meaningful contributions to waste disposal is measured in decades.[17] Therefore, unless the total system can be implemented and sustained reliably for several decades, the waste-reduction benefits are significantly diminished because the large quantities of actinides and fission products remaining would still need to be managed if continued operation broke down at any point in the cycle. Organizations, rules and regulations, assignments of responsibility, and funding arrangements would need to be established in ways that would provide reasonable assurance of the necessary stability over many decades.

Summary

The sustained commitment needed to develop and demonstrate the technologies and to build and operate the succession of required major facilities of an S&T system would represent institutional complexity and magnitude unprecedented in the civilian sector. Because of the required commitment of funding and the technical and financial risk entailed, it appears that it would be necessary for the U.S. government to accept the lead management and financial responsibility. A cohesive national intent would have to be established, a tightly managed development program organized, and the funds for effective implementation regularly provided. The last two decades of the U.S. government-led nuclear programs provide little confidence that such conditions can be established and maintained.

REGULATORY ISSUES

This section summarizes the status of the regulations applicable to the major steps in the once-through fuel cycle

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[17] The impact of waste forms and repository loading would be seen earlier.

and the handling of the resulting waste, as well as the possible applicability of current regulations to the additional facilities and steps required by an S&T fuel cycle.

Current Federal Regulations

Once-Through Cycle

The various aspects of the once-through cycle are subject to several of the titles of the U.S. Code of Federal Regulations. The applicable portions of the code are noted in the text below.

Uranium Mining. No license from the Nuclear Regulatory Commission (NRC) is required for the mining of uranium. However, uranium mines are subject to a variety of other laws. The Federal Water Pollution Control Act (FWPCA) (USC 33, 1251-1387) directs the Environmental Protection Agency (EPA) to set effluent limitations and new source performance standards for uranium mines (40CFR440.34). Under the Clean Air Act (CAA), the EPA has issued a national emission standard for ^{222}Rn emissions from underground uranium mines (40CFR61.20-61.26) and performance standards for metallic-mineral processing plants, which include open-pit uranium mining operations (40CFR60.380-60.386). The Safe Drinking Water Act (SDWA) (USC 42 300f-330j-26) would apply as well if a mine were contaminating public drinking-water supplies (USC 42 300i[a]; Scott, 1984).

Uranium Milling. Either the NRC or the so-called "agreement states" license uranium mills.[18] The regulations cover design, siting, environmental monitoring and planning, finances, inspections, operations, shutdowns, tailings, and decommissioning (10CFR40). NRC licensing of a mill may require the filing of an environmental impact statement (CFR 10 51.20[b][8]).

The NRC's Standards for Protection Against Radiation (10CFR20)[19] (commonly called the NRC radiation protection standards) apply to milling operations (10CFR20.2). These regulations protect employees and offsite individuals by setting limits on permissible radiation exposure levels (10CFR20.101/20.105) and on radioactivity in effluents (10CFR20.106). The regulations also direct NRC licensees to "make every reasonable effort to maintain radiation exposures, and releases of radioactive materials in effluent to unrestricted areas, 'As Low As Reasonably Achievable (ALARA)'" (10CFR20.1[c]). The ALARA concept is a principal foundation of NRC regulation (see Bremberg, 1989).

EPA's environmental radiation protection standards for nuclear power operations (40CFR190)[20] (EPA nuclear fuel-cycle standards) also govern uranium milling. In addition, EPA has adopted standards for radionuclide emissions under the CAA (the EPA radionuclide standards) that govern mills. However, the 1990 CAA amendments provide that EPA may rescind its authority over mills and other fuel-cycle facilities if it determines that regulation by the NRC "provides an ample margin of safety to protect the public health" (USC 42 7412[d][9]).[21]

EPA also regulates mills under the FWPCA. Each mill must obtain a permit under the National Pollutant Discharge Elimination System (NPDES) (40CFR125). However, in 1976 the Supreme Court held that pollutants subject to the FWPCA do not include "by-product," "source," or "special nuclear" material regulated by DOE and NRC under the Atomic Energy Act (AEA) of 1984 (*Train v. Colorado Public Interest Research Group*, 1976). Therefore, an NPDES permit limits only nonradioactive mill effluents.

Conversion. Conversion facilities are subject to many of the same requirements as uranium mills. The NRC or agreement states license conversion plants pursuant to the regulations for source material (10CFR40) and special nuclear material (10CFR70). These regulations cover, for example, effluent monitoring, physical security and contingency plans, materials accounting and control, decommissioning, prevention of accidents, and/or protection if accidents occur. The NRC radiation protection standards (10CFR20) apply, as do the EPA nuclear fuel-cycle standards (40CFR190) and EPA radionuclide standards (40CFR61).

Enrichment. Existing DOE enrichment facilities have historically been exempt from NRC licensing requirements (10CFR40.11, 50.11, 70.11). Instead, they had to comply with DOE orders and EPA requirements. However, with the establishment of the U.S. Enrichment Corporation, the future operation of these plants will be regulated by the NRC as well as the EPA and the Occupational Health and Safety Administration under rules promulgated in 10CFR76. Legacy issues will continue to be regulated under DOE orders and EPA requirements.

Planned commercial enrichment facilities, such as the one proposed by the Louisiana Energy Services partnership, would originally have had to meet the NRC licensing requirements for production and utilization facilities, including

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[18] An agreement state as designated in an NRC regulation means any state with which the NRC has entered into an effective agreement under subsection 274b of the Atomic Energy Act of 1954 to carry out certain aspects of the regulation of radiation.

[19] The standards were substantially revised in 1991. See 56 *Federal Register* 23360 (1991).

[20] These were issued pursuant to EPA's Atomic Energy Act authority derived from Reorganization Plan No. 3 of 1970, U.S. Code Title 5, Appendix 1.

[21] See also 56 *Federal Register* 18735 (1991).

reactors (10CFR50). Under these requirements, the facility would have had to first obtain a construction permit and then a license to operate the facility (10CFR50.22-50.23). However, in 1990, Congress authorized the NRC to issue a combined construction permit and operating license for uranium enrichment and separation facilities (PL101-575). The NRC has developed design criteria and regulations for enrichment facilities (53 *Federal Register*, 1988; 56 *Federal Register*, 1991), and the 1990 legislation mandates that an environmental impact statement be submitted and an adjudicatory hearing be held. Commercial enrichment facilities must also comply with NRC radiation protection standards (10CFR20), EPA nuclear fuel-cycle standards (40CFR190), and EPA radionuclide standards (40CFR61).

Fuel Fabrication. Commercial fuel fabrication facilities are subject to NRC licensing regulations for source material (10CFR40) and special nuclear material (10CFR70). They are also subject to NRC radiation protection standards (10CFR20), EPA nuclear fuel-cycle standards (40CFR190), and EPA radionuclide standards (40CFR61).

Licensing of Nuclear Power Plant Operations. The AEA of 1984 established a two-step licensing process for commercial nuclear power plants, whereby a utility applies for a construction permit (at which time a public hearing is required) and then, after plant construction, seeks an operating license (subject to another public hearing upon request of an interested party) (USC 42 2235/2239(a)). In 1989, however, after several years of unsuccessful attempts to convince Congress to amend the AEA to provide for a one-step process, the NRC adopted regulations (10CFR52) providing for a "combined license" (10CFR52; 54 *Federal Register*, 1989). Under this procedure, the NRC may issue a construction permit with a conditional operating license (10CFR52.97). After completion of the plant, and upon an NRC finding of conformity with the "acceptance criteria" specified in the combined license, the NRC can authorize plant operation (10CFR52.103).

This new licensing process revises the current approach in two ways. First, it requires that most of the issues previously raised in operating license hearings be scrutinized early in the licensing process at the time of the application for the construction permit. For example, the adequacy of emergency planning (10CFR50.47; 54 *Federal Register* , 1989) and whether the facility will be operated in conformity with the AEA must now be considered up front (10CFR52.97). Second, the new regulations diminish the opportunity to apply for a postconstruction hearing. For example, they would not provide the right to a hearing concerning new safety information that comes to light during construction and raises new questions about the plant's conformity with the AEA (10CFR52.103). The full Court of Appeals for the District of Columbia Circuit recently upheld the legality of the new NRC regulations after they had been vacated, in part, by a three-judge panel.

Once licensed and operating, a nuclear power plant is subject to an array of regulations governing operations (10CFR50.54), inspections (10CFR50.70), recordkeeping (10CFR50.71), emergency notification (10CFR50.72), reporting (10CFR50.73), physical protection of facilities and nuclear materials (10CFR73), material accounting and control (10CFR75), and financial protection for accidents and decommissioning (10CFR140). (A power plant license includes related operational instructions, including "limiting conditions for operation" that require shutdown under specified circumstances.) Once a year the nuclear utility must report offsite dose calculations that demonstrate compliance with the EPA nuclear fuel-cycle standards, and twice a year a facility must report effluent quantities (10CFR190; 56 *Federal Register*, 1991).

Separations/Reprocessing. The AEA defines a "production facility," in part, as "any equipment or device determined by rule of the Commission to be capable of the production of special nuclear material in such quantity as to be of significance to the common defense and security, or in such manner as to affect the health and safety of the public …" (AEA, Section 11[v]). On its face, this definition does not appear to encompass reprocessing facilities. However, in implementing regulations the NRC has defined production facility expansively to include not only a facility that produces special nuclear material anew but also one that separates existing special nuclear material from other substances, i.e., reprocessing. NRC regulations state that "any facility designed or used for the separation of isotopes of uranium or … plutonium, except for laboratory-scale facilities" constitutes a "production facility" requiring licensing of construction and operation (10CFR50.2). Such a facility would be licensed under 10CFR50, the standard licensing regulations for nuclear power facilities.

A reprocessing facility may not be eligible for a combined license under the 10CFR52 regulations for nuclear power plants adopted by the NRC in 1989. However, a reprocessing facility might, nevertheless, still be eligible for a combined license if it fell within the exception created by Congress in 1990 for plants "capable of separating the isotopes of uranium or enriching uranium in the isotope 235" (AEA, 11[v]; USC 42 2014[v]) (see previous discussion under *Enrichment*).

A separations and reprocessing plant would be subject to NRC radiation protection standards (10CFR20) as well as the EPA nuclear fuel-cycle standards (40CFR190) to the extent that reprocessing or separations "directly support[s] the production of electrical power for public use utilizing nuclear energy. …" (40CFR190.02[b]).

Transmutation by Reactor. NRC regulations define a "utilization facility" subject to NRC licensing as "any nuclear reactor other than one designed or used primarily for the formation of plutonium or ^{233}U" (10CFR50.2). The Energy Reorganization Act of 1974 provided that demonstration liquid-metal fast breeder reactors and any other new "demonstration nuclear reactors" were subject to NRC licensing "when operated as part of the power generation facilities of an electric utility system, or when operated in any other manner for the purpose of demonstrating the suitability for commercial application of such [reactors]."

Transmutation by Accelerator. Preliminary research and discussions with NRC and congressional committee lawyers have not given the committee a clear answer as to whether, under existing law, a license would be required for the construction and operation of an accelerator used to transmute waste.

The AEA requires a license for the construction and operation of any "production" or "utilization" facility with the exception of "facilities under contract with and for the account of" DOE (AEA, section 100). The AEA definition of a production facility appears above; NRC regulations define a production facility, in part, as follows:

> Any facility designed or used for the processing of irradiated materials containing special nuclear material, except (i) laboratory-scale facilities designed or used for experimental or analytical purposes, (ii) facilities in which the only special nuclear materials contained in the irradiated material to be processed are uranium enriched in the isotope U-235 and plutonium produced by the irradiation, if the material processed contains not more than 10^{-6} grams of plutonium per gram of U-235 and has fission product activity not in excess of 0.25 millicuries of fission products per gram of U-235; and (iii) facilities in which processing is conducted pursuant to a license issued under Parts 30 and 70 of this chapter, or equivalent regulations of an Agreement State, for the receipt, possession, use, and transfer of irradiated special nuclear material (10CFR50.2).

In discussing the difference between the previous definitions discussed in the above statutory and regulatory definitions, the key to whether an accelerator transmuting waste would be considered a production facility for licensing purposes was likely to be (1) whether special nuclear material or some other fissionable isotope would be produced by the accelerator or (2) whether neutron bombardment could somehow cause additional special nuclear material to be separated from the material being processed. If neither of these questions could be answered affirmatively, an accelerator transmuting waste would not constitute a "production facility" subject to NRC licensing under Part 50. Even though the regulatory definition looked as though it might encompass accelerators, it had been written more expansively than the statutory definition in order to encompass reprocessing facilities that, although they do not produce special nuclear material anew, do separate it into a usable form and therefore could be said to "produce" it.

The AEA defines a utilization facility as "any equipment or device, except an atomic weapon, determined by rule of the Commission to be capable of making use of special nuclear material in such quantity as to be of significance to the common defense and security, or in such manner as to affect the health and safety of the public, or peculiarly adapted for making use of atomic energy in such quantity as to be of significance to the common defense and security, or in such manner as to affect the health and safety of the public …." NRC regulations, however, currently limit the definition of a utilization facility to "any nuclear reactor other than one designed or used primarily for the formation of plutonium or ^{233}U."

Whether an accelerator used to transmute waste would be considered a utilization facility hinges on whether neutron bombardment of the waste material would produce heat used to generate power. If it would not—even if heat were produced elsewhere in the accelerator transmutation system and were used to produce power—the accelerator would not constitute a utilization facility.

Even though an accelerator might not, under current law, be deemed either a production or utilization facility, it would still most likely require a materials license under 10CFR70. This license might have to be issued by NRC if the quantities of material to be handled at the facility would be greater than critical mass. Otherwise, it could be issued by an agreement state.

Congress might also legislate a requirement that an NRC license be obtained for the construction and operation of an accelerator used to transmute waste. In the Energy Reorganization Act of 1974, for example, Congress provided NRC with licensing and regulating authority over liquid-metal fast breeder reactors, other new demonstration reactors, facilities used to store HLW resulting from NRC-licensed activities, and facilities used for long-term storage of defense HLW.

Waste Storage and Disposal

This section discusses the regulatory aspects of storing spent nuclear fuels and disposing of nuclear fuel-cycle wastes.

Spent-Fuel Storage (pool). The vast majority of civilian spent nuclear fuel is stored in pools at reactors. The NRC licenses the construction and operation of these fuel pools as part of the normal (e.g., 10CFR50) licensing process for reactors. An independent fuel pool would be licensed un der 10CFR72 (Licensing Requirements for the Independent Storage of Spent Nuclear Fuel and High-Level Waste).

Spent-Fuel Storage (dry cask). DOE is seeking to site and construct a monitored retrievable storage facility for the receipt and storage of spent nuclear fuel from NRC-licensed reactors. Under the Energy Reorganization Act, section 202(3), such a facility would be subject to NRC licensing under 10CFR72. The same regulations would apply to development of dry-cask storage at an S&T facility built in conjunction with S&T facilities.

While EPA has developed standards for the protection of the environment from stored high-level and TRU wastes, the standards are currently written to apply to such wastes only when at a repository site (40CFR191).

Geologic Repository (high-level and TRU Wastes). Development and operation of one or more repositories for the disposal of HLW are governed by the Nuclear Waste Policy Act (NWPA). The law, as amended in 1987, directs DOE to investigate the site identified at Yucca Mountain, Nevada, for the possible development of a repository. After site investigation, the Secretary of Energy may recommend approval of the site to the President, who, in turn, may make a formal recommendation of approval to Congress. While Nevada may then formally veto this determination, the law provides that Congress can override the state's decision.

If site approval is sustained by Congress, the NRC would consider DOE's plans for the development and operation of the repository in a formal licensing process. The licensing process would be governed by specific NRC regulations (40CFR191). The NRC requirements are dictated, to a large extent, by standards developed by EPA in 1985 setting limits on releases of radioactivity to the general environment, including groundwater, and on human exposure (40CFR191). The First Circuit Court of Appeals vacated the standards in 1987 because of inconsistencies with the Safe Drinking Water Act in the protection afforded by the standards to underground sources of drinking water and inadequacies in the standard governing individual exposures. Five years after the court's decision, EPA has not succeeded in issuing revised standards for a repository that might be established at Yucca Mountain because of several controversial issues. These include, for example, the release limit for ^{14}C, how inadvertent human intrusion should be analyzed, and the level of certainty required for a determination of compliance. In 1992, Congress directed a National Research Council study to provide recommendations to the EPA concerning the technical basis for health-based standards. Details are given in Chapter 2.

Low-Level Waste Disposal. Low-level wastes from fuel-cycle facilities are generally regulated by NRC and agreement states under 10CFR61. These regulations divide LLW into three classes, depending on radioactivity and half-lives (10CFR61.10-61.31). They also set out detailed requirements for licensing LLW disposal sites. However, NRC does not usually regulate accelerator-produced radioactive waste (activation products, etc.) because such waste generally does not meet the definition of "by-product material" under the AEA, that is, "radioactive material … yielded in or made radioactive by exposure to the radiation incident to the process of *producing* or *utilizing* special nuclear material … "(USC 42 6901-6992k).

If it were determined that the accelerator itself constituted a production or utilization facility under the AEA, any accelerator-produced waste would constitute by-product material. If it were agreed that the accelerator were not such a facility, then the material would be exempt from NRC regulation. EPA has proposed to regulate accelerator-produced wastes but has not succeeded in developing final regulations. (A handful of states have developed regulations for non-AEA wastes, but these do not appear to cover accelerator-produced wastes.)

The Regulatory Role of the States

Any assessment of the impact of environmental statutes and regulations must also take into account the increasingly important role of state governments. For example, state and local requirements govern several facets of uranium-mining operations, including land disturbance and groundwater pollution caused by uranium exploration activities and mined land reclamation. With the new federalism of the 1980s, many of the states have assumed leadership roles not only in the development of their own environmental statutes and regulations, but also in the implementation of the major federal environmental laws. In fact, through the process of EPA state agreements, the majority of the states have been delegated the authority for the permitting and enforcement activities for the major federal laws, including the Resource Conservation and Recovery Act, the Clean Water Act, and the Clean Air Act.

In addition to the federal statutes, laws and regulatory requirements that could affect the siting and operation of DOE waste-management facilities have also been developed by the majority of states. These approaches may parallel federal statutes but often include additional requirements concerning waste storage and repository siting; worker and community "right-to-know" waste tracking; air, water, and waste characteristics, quantities, and radioactivity limitations; and reporting. An inventory of all such state and local regulatory requirements is beyond the scope of this report; however, an evaluation of these additional requirements is an essential prerequisite for the development of any technology for waste treatment or storage.

The tripartite agreements developed at several DOE fa cilities illustrate the current trend toward shared decision-making. The increased involvement of the states in waste management decisions brings with it an increasing need to address public concerns about risks. Public opinion on nuclear waste and other environmental issues has had a profound impact on the political process on the state and local levels. Elected officials must be responsive to local concerns about environmental and health risks, so state policies are more likely than federal statutes to require extensive evaluation and control of actual and perceived risks.

Conclusions

If one accepts the eventual need for reprocessing and recycling as a part of a sustainable supply of nuclear power, then challenging regulatory and licensing issues will inevitably have to be faced. For the most part, the fundamental federal regulatory framework needed to license the facilities required to implement S&T technology exists. Most of the fuel-cycle operations involved in S&T would be governed by federal regulations that are established and familiar.

There are two notable exceptions to the preceding statement, however. First, the applicability of existing regulations to accelerator-based transmutation devices with closely coupled continuous fuel reprocessing is very unclear. Second, one or more major generic environmental impact statements would be required before reprocessing could begin. In addition, state and local governments are becoming increasingly involved in regulatory activities, both by enforcing federal regulations and by applying additional requirements. This introduces additional uncertainties into the future regulatory environment for S&T facility deployment. For instance, neither the underlying standard nor the regulations relevant to a potential repository at Yucca Mountain is final. It is very difficult to predict what the regulatory and licensing situation will be at the time when S&T technology might be implemented. If the major changes in licensing and regulation experienced over the last 15 years are a harbinger of the future, the situation is likely to be radically different from what exists now.

RESEARCH AND DEVELOPMENT NEEDS

This report addresses application of separations and transmutation (S&T) technology to two distinct waste streams: civilian reactor spent fuels and neutralized defense high-level waste. At present, transmutation technology is not developed to the point at which it could be applied immediately and directly to either waste stream. Separations technology is better developed, but important advances are still possible with further research and development. As a consequence, if S&T is to be implemented, a research and development program will be required to provide the needed technology.

The committee is of the view that a sustained, carefully focused program of research and development emphasizing advanced separation technologies should provide a sound basis for future decisions on nuclear waste S&T. Following successful laboratory-scale development, engineering pilot-scale demonstration would be necessary.

Civilian Reactor Spent Fuels

It should be recognized at the outset that S&T (i.e., enhanced recovery and destruction of selected radionuclides) is inextricably intertwined with standard spent-fuel reprocessing. As a result, the discussion of research and development needs involves substantial aspects of current reprocessing technology.

There is no immediate need to deploy S&T technology—primarily because the long duration of the repository project and the economics of reprocessing are unfavorable in the current era of low-cost uranium and enrichment—so a research and development program cannot be viewed as urgent. For the near future in the United States, S&T is best regarded as a contingency option. (Present U.S. policy continues to hold that spent reactor fuel will not be reprocessed.) On the other hand, implementation of reprocessing/S&T might become desirable under a variety of situations, ranging from a change in the economic viability of actinide recycling to being required to facilitate establishment of a repository, to ameliorating the climatic impacts of other energy production technologies. Because the time required to develop S&T technology is long compared to the time over which we can foresee the degree to which it would be needed, it is desirable to continue to develop S&T technology until such time as a firm determination of need or the lack thereof can be made. Justifications for sustaining a modest level of research and development on S&T technology include:

- the inefficiency of ceasing research and development now, and then restarting it as a part of a crash program in the future;
- the desirability of maintaining a base program on fast reactors and actinide recycling in the United States, so that we will have the expertise and credibility required to be effectively involved in international nonproliferation activities and also have access to related technology developments in other countries; and
- the need to develop improved technologies for transmuters and reprocessing so as to improve safety, economics, and proliferation resistance if and when needed.

Continued research and development in S&T will pro vide a basis for making sound decisions at a later time when S&T might appear to merit reconsideration.

In general, continuous processes (e.g., solvent extraction) are to be preferred over semicontinuous processes (e.g., ion exchange), which in turn are preferred over batch processes (e.g., electrorefining) because of increased efficiency and reliability. Inorganic reagents are preferable to organic reagents because of their greater stability, although waste volume needs to be taken into account if the ion exchange medium becomes a waste because of degraded performance.

As implied in the earlier discussion of priorities, the highest priority for transmuters clearly belongs to LWRs for the initial phases of S&T, followed by LMRs (possibly with some LWR component) for the longer-term transmutation of actinides. Other transmuters do not appear to offer advantages commensurate with the development risks and costs, although limited support of these devices for S&T purposes might be warranted if the base development program were to be funded for other reasons.

The committee believes that research and development activities should remain at the minimum scale consistent with maintaining the ability to obtain results needed for technology assessment and for expeditious implementation in an operating facility. In general, this minimum scale will involve a sequence of activities at the cold bench, hot bench, cold engineering (or pilot plant), and hot engineering (or pilot plant) scales. If demonstration at existing facilities or pilot-scale work will not suffice, transmuters may require an operating demonstration facility and technology assessment to reduce the number of demonstrations to a minimum. However, it is to be emphasized that use of actual feed materials is imperative in the research and development phase.

Neutralized Defense High-Level Waste

In the case of neutralized defense HLW, the forces driving S&T research and development are very different from those related to spent reactor fuel. These diluted wastes contain small amounts of long-lived species relative to those in spent fuel, and the volume of the wastes is very large. The objective for defense wastes is the use of S&T technology to reduce the volume of HLW by processing the waste into as small a volume of HLW and as large a volume of LLW as are economically feasible within the balance between separations and disposal costs.

These wastes are primarily composed of common chemicals that do not need to be treated as HLW (i.e., vitrified and sent to a repository). The removal of these chemicals could reduce the volume of HLW by factors of 5 to 200, saving billions of dollars with little change in risk. Tank-waste remediation is already beginning at Hanford and Savannah River, with existing technologies, but the remediation programs will take decades and therefore could benefit from a parallel program of research and development.

The committee notes that research and development for defense HLW should not include transmutation of long-lived radionuclides. The amount of radionuclides relative to that in LWR spent fuel is simply too small to justify such an effort.

Research and development for Handford, Savannah River, and Oak Ridge can be coordinated and shared, and the longer-term research component should benefit all those sites.

In general, only aqueous processes should be considered for processing defense HLW. The range and merit of the various alternative processes are discussed in detail in Chapter 3. While small, specific problems may require other classes of processes, the genesis and composition of these wastes dictate an aqueous approach (with, perhaps, the exception of the wastes at Idaho National Engineering Laboratory). As with spent-fuel processing, continuous processes are to be preferred over semicontinuous, which in turn are preferable to batch processes. In any case, it is imperative that the normal technology development sequence (cold/hot bench, cold/hot pilot plant) be followed, except in cases where there are clear and immediate safety hazards or severe deterioration of containment. To do otherwise entails large economic and credibility risks that should be viewed as unacceptable.

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Appendixes

Appendix A

Statement Of Work

The National Research Council Board on Radioactive Waste Management, in conjunction with the Board on Chemical Science and Technology and the Energy and Engineering Board, will arrange for the nomination of a multidisciplinary committee of approximately 18 experts in appropriate fields such as nuclear chemical and fuel-cycle engineering, nuclear engineering, separations science, accelerator physics, waste (radioactive and hazardous) disposal technologies, exposure assessment, economics, public policy, regulatory policy/procedures, nuclear reactor safety, and plant operations. The committee will meet approximately four times per year. Meetings will be conducted in accordance with National Research Council meeting procedures.

The committee will initiate its activities by conducting an issue-oriented workshop and will then broadly review the applications of separations and subsequent processing system technologies to the U.S. Department of Energy (DOE) waste management programs in the context of the National Energy Strategy. Mature technologies will be considered and, in addition, those developed sufficiently to be technically credible. Benefits and detriments to disposal of wastes will be evaluated. The review will emphasize:

- The extent to which the technologies have been demonstrated and on what scale.
- The potential benefits and detriments to disposal of high-level radioactive wastes in geologic repositories.
- The technical, scientific, economic, and regulatory uncertainties associated with those technologies, of foreign and/or domestic origin, that may have been demonstrated to only a limited degree.
- The time required for technology demonstration and its relationship to repository program timing.
- The impacts, positive and negative, of technologies on waste arisings over the entire fuel cycle.

The preceding evaluations will involve identification and discussion of the amount and chemical characteristics of the products, including secondary waste streams, of the facilities constructed to enable the technologies to be effectively applied to the waste management system. Mass balances will be developed for all significant materials entering into or deriving from the affected facilities.

The committee will evaluate the probable costs associated with the development of the various technologies through their demonstration stages, and the probable costs of the facilities and operations that would apply the technologies to the waste management system. The committee will also address other factors that might affect whether facilities applying the technologies reviewed will be constructed and operated.

A final report summarizing the committee's findings in areas reviewed will be prepared 36 months after task approval. Interim reports will be provided as appropriate. All reports will be reviewed and distributed in accordance with standing procedures and policies of the National Research Council. DOE will receive 150 copies of each report; additional copies will be provided to the National Research Council committee members and other parties in accordance with National Research Council policy. Committee reports will be made available to the public without restriction.

June 27, 1991

Appendix B

List Of Committee and Subcommittee Activities

Briefings

- Department of Energy
- Nuclear Regulatory Commission
- Environmental Protection Agency
- General Electric Company
- Electric Power Research Institute
- Argonne National Laboratory
- Los Alamos National Laboratory
- Brookhaven National Laboratory
- Westinghouse Hanford Company
- Yucca Mountain Project Office

Site Visits by Committee Members

- Argonne National Laboratory
- Los Alamos National Laboratory
- Savannah River Site
- Westinghouse Hanford Company
- THORP, Sellafield, United Kingdom
- Brookhaven National Laboratory
- Japan

Meetings

- International Workshop January 13-14, 1992, Washington, D.C.
- Committee Meetings

Meeting No. 1, October 10-11, 1991, Washington, D.C.
Meeting No. 2, January 15, 1992, Washington, D.C.
Meeting No. 3, April 9-10, 1992, Washington, D.C.
Meeting No. 4, June 25-26, 1992, Washington, D.C.
Meeting No. 5, August 18-20, 1992, Woods Hole, Mass.
Meeting No. 6, October 6-7, 1992, Washington, D.C.
Meeting No. 7, December 14-15, 1992, Irvine, Calif.

Meeting No. 8, April 14-15, 1993, Washington, D.C.
Meeting No. 9, August 4-5, 1993, Washington, D.C.

- Subcommittee on Separations

Meeting No. 1, November 12, 1991, Washington, D.C.
Meeting No. 2, January 13, 1992, Washington, D.C.
Meeting No. 3, February 10, 1992, Washington, D.C.
Meeting No. 4, April 4, 1992, San Francisco, Calif.
Meeting No. 5, August 17, 1992, Woods Hole, Mass.
Meeting No. 6, May 19, 1993, Richland, Wash.

- Subcommittee on Transmutation

Meeting No. 1, November 16, 1991, Berkeley, Calif.
Meeting No. 2, December 6, 1991, Cambridge, Mass.
Meeting No. 3, March 12-13, 1992, Idaho Falls, Idaho
Meeting No. 4, April 21-22, 1992, Los Alamos, N.M.
Meeting No. 5, July 16-17, 1992, Norwich, Vermont
Meeting No. 6, August 17, 1992, Woods Hole, Mass.
Meeting No. 7, November 1992, Berkeley, Calif.
Meeting No. 8, March 8-9, 1993, Washington, D.C.

- Subcommittee on Integration

Meeting No. 1, December 9, 1991, Washington, D.C.
Meeting No. 2, January 15-16, 1992, Washington, D.C.
Meeting No. 3, March 19-20, 1992, Washington, D.C.

Data Collection: Data on current international activities were collected through the international workshop held in January 13-14, 1992, through representatives of the United Kingdom, Germany, Sweden, France, Japan, countries of the former Soviet Union, Czechoslovakia, the Organization of Economic Cooperation and Development/Nuclear Energy Agency (OECD/NEA), and the Commission of the European Communities.

Contacts with the U.N.'s International Atomic Energy Agency and the OECD/NEA were maintained for further data collection until the end of the STATs committee's study.

For current U.S. information, contributions were received from committee and subcommittee members and a wide selection of scientists and authorities concerned with the field, especially the proponents of the different separations and transmutation concepts. A general bibliography is attached as Appendix P. Personal contacts with a large number of people involved in separations and transmutation were started quite early and maintained until the end of the STATS committee's study.

Appendix C

Base Case Once-Through Uranium Fuel Cycle for Light-Water Reactor with Geological Repository*

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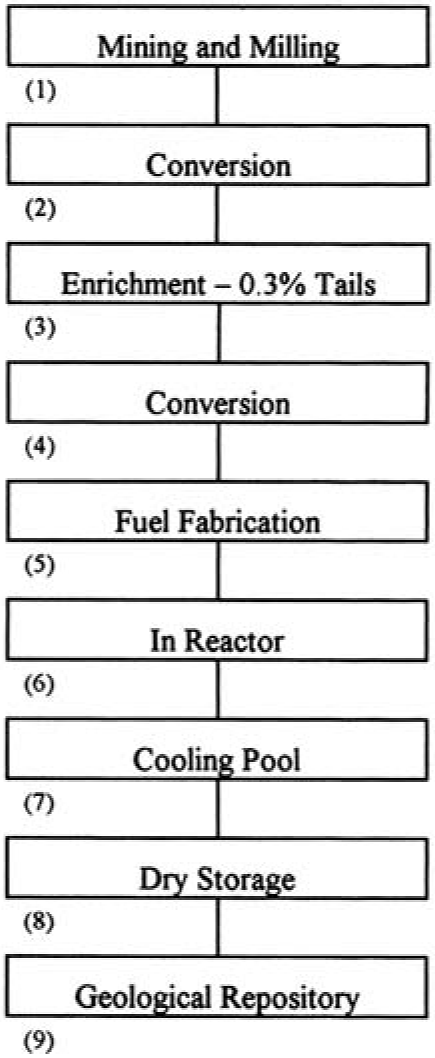
* This appendix was written by Norman Rasmussen with the Massachusetts Institute of Technology and Allen Croft with the Oakridge National Laboratory. Both are members of the Committee on Separation Technology and Transmutation Systems.

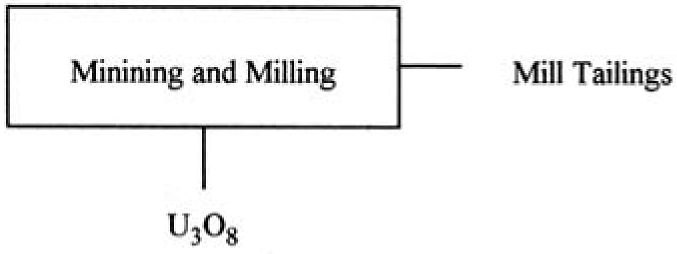
This is the flowsheet for the fueling of a 1,000 MWe light-water reactor (LWR) for 1 year of operation with a 80% capacity factor. A once-through fuel cycle is assumed. The two columns on the left show the amount of uranium at the output of each step in metric tons (1 T = 1,000 kg) and the percent enrichment of ^{235}U. The right-hand column shows the percentage of uranium lost in the process. The technical data used in this fuel cycle are taken from the textbook *Nuclear Chemical Engineering* by Benedict, Pigford, and Levi, 2nd edition, McGraw Hill, New York, 1981.



Uranium, Metric Tons (T)*	Enrichment	Loss	
193	0.71%	5%	Mining and Milling (1)
192	0.71%	0.5%	Conversion (2)
27.2	3.2%	0.5%	Enrichment – 0.3% Tails (3)
27.1	3.2%	0.5%	Conversion (4)
26.9	3.2%	0.5%	Fuel Fabrication (5)
25.9	0.83%	0	In Reactor (6)
25.9	0.83%	0	Cooling Pool (7)
25.9	0.83%	0	Dry Storage (8)
25.9	0.83%	0	Geological Repository (9)

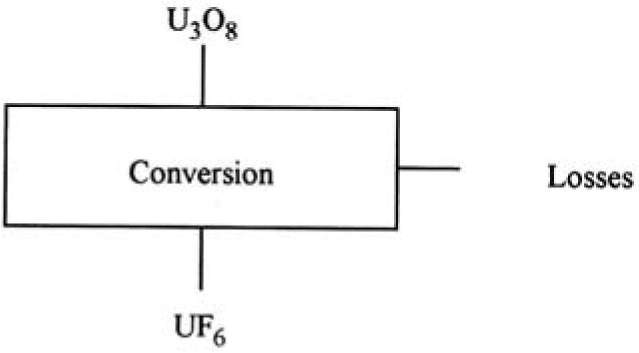
* Start 202 in Ore

STEP 1



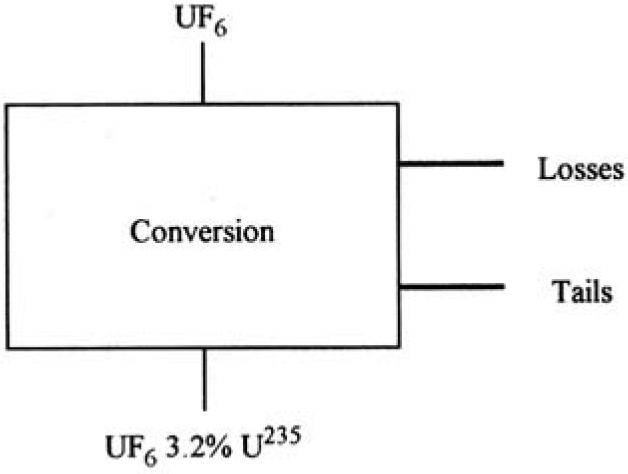
Description:	In this step, the mined ore is processed to concentrate the uranium. A combination of methods is used including leaching, precipitation, solvent extraction, and ion exchange. The output stream is U_3O_8. The main byproduct stream is mill tailings that contain a small fraction of the initial uranium and all its initial radioactive daughter products.
Input:	The input is the mined ore and the amount depends on the grade of the ore, typically around 0.15–0.2%.
Output:	228 T of U_3O_8.
Environment:	The principal impact is due to disposal of mill tailings that can be dispersed by rain and wind and produce a significant dose to the public.
Siting Issues:	The ore processing activity can be noisy and dusty. It is usually carried out near the mine and removed from population.
Public Health:	This process is a predominant contributor to occupational dose and dose to the public.

STEP 2



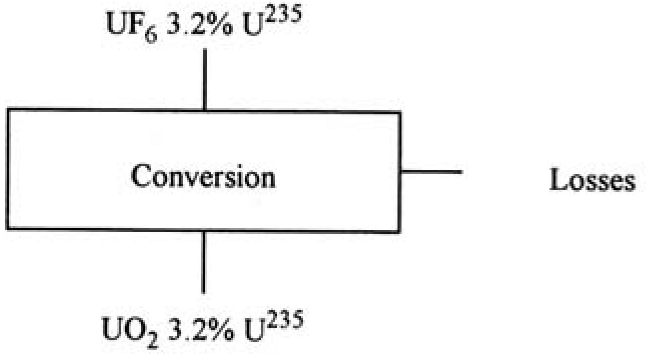
Description:	In this step, the U_3O_8 is chemically processed to convert it into UF_6, a gas that is the input for the gaseous diffusion process.
Input:	228 T of U_3O_8.
Output:	284 T of UF_6
Environment:	There is very little impact, and it is mostly associated with uranium losses.
Siting Issues:	This process is like many chemical plants. It would be restricted to an industrial zone. Uranium separated from its daughter products is only weakly radioactive.
Public Health:	This process would be a small contributor to the cycle's public health effects.

STEP 3



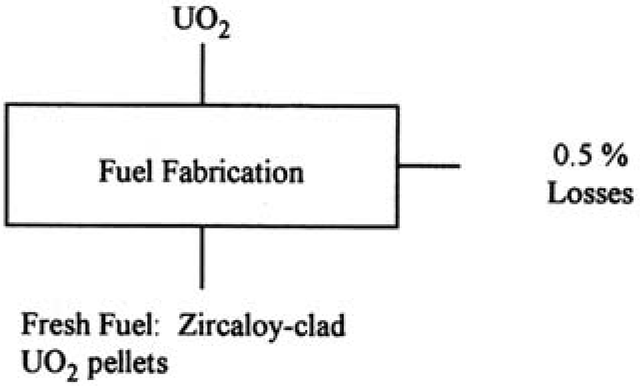
Description:	In Step 3, the natural uranium UF_6 is put through the enrichment plant to increase the percentage of ^{235}U from 0.71 to 3.2 at a tails assay of 0.3%.
Input:	284 T UF_6 (200 T uranium 0.71% ^{235}U).
Output:	40.1 T of UF_6 product (27.2 T uranium 3.2% ^{235}U). 242.4 T UF_6 tails (164 T uranium 0.3% ^{235}U).
Losses:	0.5% (1 Ton natural uranium).
Environment:	The enrichment plant is very large and uses moderate-to-large amounts of electricity (depending on the technology). Uranium losses lead to a small amount of environmental degradation. The tails stream constitutes a major output stream, the disposition of which has not been decided.
Siting Issues:	A large nonurban site is needed.
Public Health:	There are some occupational health issues—small public health concerns.

STEP 4



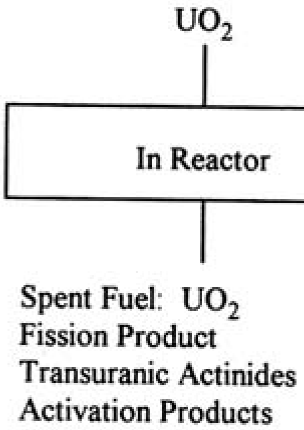
Description:	The enriched UF_6 must now be converted to UO_2, which is the material used in reactor fuel elements. This is a chemical process.
Input:	27.2 T uranium 3.2% ^{235}U as UF_6.
Output:	27.1 T uranium 3.2% ^{235}U as UO_2.
Losses:	0.5% uranium.
Environment:	There is very little impact, and it is mostly associated with uranium losses.
Siting Issues:	The process is like many chemical plants. It would be restricted to an industrial zone. Uranium separated from its daughter products is only weakly radioactive.
Public Health:	This process would be a small contributor to the cycle's public health effects.

STEP 5



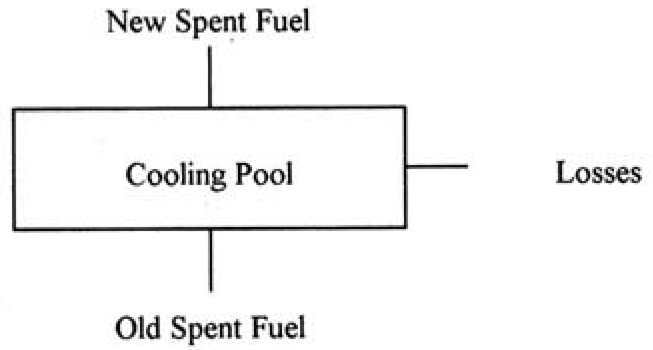
Description:	In this step, the UO_2 is compacted into pellets (0.3 to 0.4 inches diameter x 0.5 inches long), and these pellets are sutured and placed into Zircaloy tubes about 12 feet long. These tubes are assembled in square arrays of about 17×17 tubes per fuel element.
Input:	27.1 T uranium 3.2% ^{235}U as UO_2.
Output:	26.9 T uranium 3.2% ^{235}U as UO_2.
Losses:	0.5% uranium.
Environment:	There is very little impact, and it is mostly associated with uranium losses.
Siting Issues:	The process is like many chemical plants. It would be restricted to an industrial zone. Uranium separated from its daughter products is only weakly radioactive.
Public Health:	This process would be a small contributor to the cycle's public health effects.
Other Issues:	Uranium in this step is well below the enrichment useful for nuclear weapons production.

STEP 6



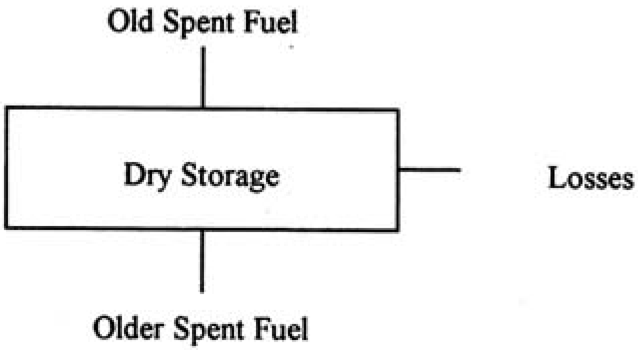
Description:	The fabricated fuel is put in the reactor and used to sustain the chain reaction. It is typically irivaluated for about 33,000MWd per T of uranium. The enrichment at the end of the run of 1,100 days is about 0.83% ^{235}U.
Input:	26.9 T uranium 3.2% ^{235}U as UO_2.
Output:	25.9 T uranium 0.83% ^{235}U as UO_2.
By-products:	See enclosed tables giving concentrations of major radionuclides in spent pressurized water reactor fuel (Appendix A. 1, ORNL/TM-7431) in terms of curies, watts, and ingestion hazard as a function of time. Tritium concentrations are about 22,000 Ci/yr (818 Ci/MTHM). About two-thirds is from ternary fission. About one-third is from an assumed 2 ppm of lithium in the fuel, but this number is speculative. See attached sheet for the ^{14}C inventories in the fuel and their distribution (ORNL/TM-6051).
Environment:	During operation, all reactors release small amounts of radioactivity, including tritium, noble gasses, and ^{14}C.
Siting Issues:	Tend to be distinctive, visible, and controversial, primarily because of potential accidents.
Public Health:	Routine releases are generally small, but the worst of major accidents could result in substantial offsite doses.
Proliferation:	UO_2 input fuel has an enrichment of 3.2 ^{235}U that is well below that required for use in nuclear explosives. The output contains 0.246 T of plutonium, which is mixed with the highly radioactive fission products, so it is considered to be self-protected.

STEP 7



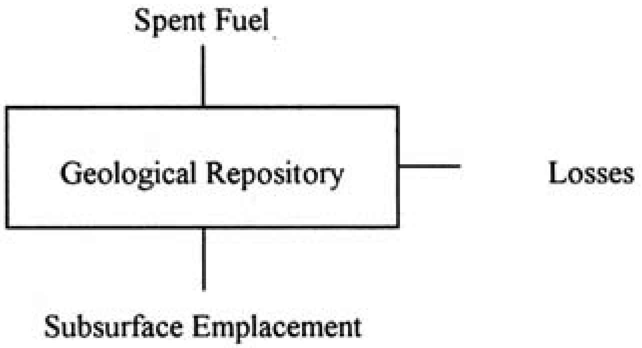
Description:	The fuel assemblies are unloaded from the reactor and stored in the reactor cooling pool to remove decay heat and provide shielding from radiation. In the United States, reprocessing of commercial fuel is not yet economic, so reprocessing is not being done. Thus, all commercial fuel at power plants is being stored in pools at the reactor site. Some fuel has remained stored in this way for over 20 years.
Input:	Spent-fuel assemblies discharged from the reactor.
Output:	Spent fuel, typically decayed for many years
Environment:	A small fraction of rods will begin to leak while in storage. Since they are under water, most leaked radioactivity will be trapped in the water. Constant monitoring will catch leaks shortly after they occur. Once identified, leaking fuel assemblies are placed in special containers maintained for this purpose.
Public Health:	There is some exposure of workers at facility. There is very little impact on the public from this operation.
Losses:	Very small.

STEP 8



Description:	Once all storage pools are full, a number of utilities are proposing to store them in special shielded storage casks on or near the surface. The elements would typically have been in cooling pools for many years and much of their radioactivity would have been reduced by decay. The casks would be designed to conduct the small amount of heat still being generated. Presumably, the fuel would be quite safe in a storage cask until it was shipped to a geologic repository.
Input:	Spent-fuel elements after significant cooling in pool.
Output:	Spent-fuel elements on their way to a repository or monitored retrievable storage facility.
Environment:	A small number of leaks might develop, but if casks are well sealed, very little loss to environment would be expected.
Public Health:	There is some exposure of workers at facility. There is very little impact on the public from this operation.
Losses:	Very small.

STEP 9



Description:	In this step, the spent fuel would be sent to the repository for final emplacement. At this point, they would be removed from the cask and possibly repackaged into a tighter configuration, or they might be sequestered in their casks. If repackaging is done, some occupational exposure would be expected along with some small release of radioactivity. Once emplaced, the container is designed to survive 1,000 years, and so no leakage from the repository should occur before that time. At times greater than 1,000 years, the container is assumed to have failed and the leakage rate is then controlled by the rate of leaching and migration to the repository boundary.
Input:	Spent fuel.
Output:	None. Permanent emplacement.
Environment:	Gradual inflow of water, waste-form degradation, radionuclide dissolution, and transport is assumed. Releases are predicted to be extremely small and to occur over extremely long times. Intrusion is possible (e.g., drilling through a waste package).
Public Health:	Impacts are predicted to be extremely small, except to an intruder.
Losses:	Loss rate is controlled by leaching and migration (inventory of radioactivity 1,000 years after emplacement).

Appendix D—

Separations Technology—Additional Information

Introduction

All current processing plants for reactor fuel have similar steps. The first is the separation of the fuel from a protective sheath called cladding. This sheath is metal; Zircaloy, aluminum, and stainless steel are the usual materials. These materials are alloys, and the nature of the minor elements must be considered. Zircaloy cladding may be made with a few percent of tin or niobium. The aluminum cladding may contain silicon, and stainless steel cladding is usually made of iron with 18% chromium and 8% nickel. Fast reactor cladding is essentially iron and chromium with very little nickel present.

In processing of commercial spent oxide fuels, Zircaloy is removed by mechanical chopping of the fuel rods into segments, followed by dissolution of the spent nuclear fuel in nitric acid. Zircaloy may also be dissolved away from metal fuel with nitric and hydrofluoric acids. In this zirconium cladding dissolution procedure, the dissolver solution is later reacted with aluminum nitrate to form fluoride ion complexes in order to prevent excessive corrosion of downstream process equipment and to supply a salting agent to promote the extraction process. This process generates large volumes of waste per unit volume of fuel processed. The mechanically removed hulls may retain some activity but are a minor process and waste volume problem.

Aluminum cladding is often dissolved in nitric acid catalyzed with mercury; alternatively it may be dissolved in sodium hydroxide sodium nitrate solution. The caustic solution is then filtered to recover the actinide fraction, or made acidic to produce the feed solution. In some processes like REDOX, the aluminum nitrate serves as a salting agent for the extraction of plutonium and uranium. Unfortunately, this aluminum nitrate constitutes a waste stream that is ten times larger than the fission products themselves.

The usual approach with stainless steel is to shear the cladding and dissolve the fuel oxide in HNO_3, leaving the steel hulls in solid form for final disposal. The stainless steel is not dissolved because the elements in the alloy interfere with the aqueous separation processes, but it can be dissolved with anodic corrosion methods when using pyrochemical separation techniques.

PUREX feed is made by leaching the fuel from the cladding, thus leaving nitric acid as the only salting agent and the actual fuel as the solute. Nitric acid is easily recycled for reuse with simple distillation and concomitant minimization of wastes. The process may be used with added salts in the feed if desired, but the optimum waste stream is not produced this way. In the case of the pyroprocesses conducted electrochemically in molten salt, the metallic sheath (usually stainless steel) might be removed from the fuel by anodic dissolution. The fuel oxide dissolves in the molten salt. If the salt is lithium chloride the oxides may be converted to the metals by reaction with lithium metal and prepared for further electrochemical processing. The oxygen present reacts to form lithium oxide, which may be removed from the salt by electrolysis with a carbon anode to regenerate the lithium metal at the cathode for reuse, and carbon dioxide which is discarded as waste. This avoids the formation of excessive solid process wastes.

Molten salts and alloys have a long history of use in the processing of nuclear weapons materials. They are used in processes for production of lithium metal and lithium compounds as well as of uranium and plutonium metals. Essentially all processes for the manufacture of fluorine and intermediate uranium compounds for uranium hexafluoride production are pyrochemical. Many of the recycle processes for uranium and plutonium, including residue recovery, are pyrochemical. There are no aqueous chemical analogues for many of the steps done by pyrochemical means, but there are advanced pyrochemical techniques that can replace many traditional aqueous processing methods.

The single-stage separation factors in pyrochemistry can be large for equilibria between liquid metal and molten salt phases, and cascades are not usually required for fuel recycle. Multistage equipment using short-stage-time centrifugal contactors originally designed for aqueous systems is being developed for those molten-salt/molten-alloy processes that require a high degree of purification. High-temperature liquid-liquid extraction equipment is similar in concept to that used for aqueous systems.

There is a large body of experience with the use of aqueous solutions and organic extraction phases for large separation cascades. There has been success using large and small separation factors, down to a stage separation factor of 1.002. This experience should be applicable to many of the proposed high-temperature molten-salt and alloy systems that generally show good separation factors for isolation of actinide elements from fission product residues.

Aqueous Processes

Some insoluble products result from high burn-up fuel dissolution in nitric acid. The solids may be complex fission-product compounds or insoluble reaction products. High-fired pure plutonium oxide does not dissolve completely. Such solids would typically be removed from the solutions by filtration or centrifugation for further treatment or discard to waste. Both of these solid–liquid separation methods can be designed for easy recycle of valuable materials. The radioactive rare gases krypton and xenon can be removed in the off-gas system, perhaps by cryogenic absorption methods. Iodine can be chemically trapped from the gas phase during dissolution. A process to separate volatile ruthenium or technetium oxides could be included at the time of initial dissolution of the spent fuel.

After dissolving the fuel, the next step is to separate the uranium and plutonium from the very radioactive fission products and the higher actinides. With the uranium in the uranyl form (+6) and the plutonium in the +4 oxidation state, these elements may be separated from all other materials of concern to almost any degree desired using a multistage cascade that yields very low losses of uranium and plutonium to waste. PUREX accomplishes this task very well, but in most plants the actual recovery of uranium and plutonium is not as good as theoretical design indicates. This is frequently due to the formulation of polymeric species during the extraction process. Solutions to this problem are now available and could be incorporated into the process.

The Butex process, based on the extraction solvent dibutoxy diethylene glycol (dibutyl carbitol) has been used in the U.K. weapons complex for the purification and isolation of plutonium. This flammable ether is used much like the "hexone" of the oxidation-reduction chemistry process (REDOX). A series cascade of a PUREX stage and a Butex stage has been used to recover and purify uranium to the part per million (ppm) level for four decades at the Y-12 Plant at Oak Ridge.

Tributyl phosphate (TBP) is used as the PUREX extractant.[1] It suffers from hydrolysis and radiolysis products that complicate the product recovery step that is carried out by stripping plutonium and uranium from the organic phase with dilute nitric acid. The solvent must be continuously purified to achieve the high recovery and decontamination levels needed. TBP is purified in PUREX plants by removing deleterious degradation products that result from both radiolytic and chemical processes; contact with sodium carbonate solution is used. Removing the degradation products from the TBP generates large volumes of contaminated wastes that must be eventually treated for disposal.

The equipment used for radiochemical separation processes needs to be designed for extreme reliability and with very low maintenance. This means that much of the chemical equipment has to be specifically designed for the operation to be performed, and generally, commercial items are not satisfactory. Much of the process equipment needed has no commercial use outside the nuclear industry.

The extraction equipment used for liquid–liquid separation processes is however, to a first approximation, independent of the solvents used. Thus, new processes that may be developed using, for example, liquid ion exchangers, phosphine oxides, phosphoric acid derivatives, or solvent systems based on amides, may entail only simple changes in operating conditions rather than new plant equipment.

In aqueous plants, extraction and stripping can be conducted in several different types of equipment. Most plants use either centrifugal contactors, mixer-settlers, or pulsed columns, occasionally some of each. Centrifugal contactors are gaining favor, because they are the most compact and fastest types of solvent extraction systems and thus minimize shielding costs, increase plant throughput, and reduce radiation damage to process reagents. Some plants employ different types of equipment in different stages of the process. To minimize radiolytic damage to solvents, equipment with short contact times (such as centrifugal contactors) may be selected for first-cycle extraction, where the greatest fission product, radioactivity, is present. Simpler equipment, such as pulsed columns or mixer-settlers, is satisfactory for second and third cycles, where radioactivity is several orders of magnitude lower.

In Europe, where there is considerable industrial experience in aqueous processes (Butex process is no longer used), many improvements have already been realized, e.g.,

- Centrifugation of fines installed.

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[1] The reactions for TBP are: (a) $UO_2(NO_3)_{2(aq)} + 2TBP_{(o)} = UO_2(NO_3)_2(TBP)_{2(o)}$; (b) $Pu(NO_3)_{4(aq)} + 2TBP_{(o)} = Pu(NO_3)_4TBP_{2(o)}$

- Plutonium recovery in UP_3 99.7%.
- TBP withstanding fuel with burn up of 4GWDT especially when using pulsed columns.
- Centrifugal contactors have already been used in second cycle operations.

Bismuth Phosphate Process

In 1941 it was known that plutonium has multiple oxidation states. A greatly expanded investigation of the aqueous chemistry of plutonium on the tracer level was initiated, which included separations methods based on precipitation, organic solvent extraction, and other approaches. It was found that plutonium coprecipitates in its reduced states with lanthanum fluoride, but not in its oxidized states. It coprecipitates with iron hydroxide and zirconium phosphate, which are typically gelatinous, hard-to-filter precipitates. A precipitation method in which most other elements would not follow plutonium was developed based on the ability of zirconium phosphate precipitate to carry plutonium in the +4 oxidation state but not in the +6 state. The reducing agent was $NaNO_2$, and the oxidizing agent was $NaBiO_3$. It was found that the addition of sulfate to uranium solutions formed complexes with the uranyl ion and suppressed precipitation of uranium phosphate, while $NaNO_2$ did not reduce $UO_2{}^{+2}$ to U^{+4}. $NaBiO_3$ oxidized Pu^{+4} to $PuO_2{}^{+2}$, and the formation of plutonyl ion prevented precipitation of plutonium phosphate from oxidized solutions. The process was soon converted to $BiPO_4$ precipitation, since it performed as well as $Zr_3(PO_4)_4$ as a carrier, and the precipitate was granular, greatly simplifying isolation. The $NaBiO_3$ oxidation step provided an internal supply of Bi^{+3} for scavenging the unwanted fission products, which were precipitated from the oxidized solutions as phosphates prior to carrying Pu^{+4} on bismuth phosphate.

This was the first large-scale radiochemical separation to exploit oxidation-reduction processes for both the purification and concentration of a single element from many other contaminants at these low concentrations. It is notable that the Bismuth Phosphate Process had a scale-up factor of 10^8 from lab to plant and produced 99.9% pure plutonium product at a 97% chemical yield, with a decontamination factor of better than 10^7 from the radioactive fission products (Lawroski, 1955). The Bismuth Phosphate Process demonstrated the practicality of obtaining decontamination factors in the 10^7 range, which was needed for manual handling of the plutonium product in later fabrication steps. The only purity difference from current processes was in the chemical yield, which was slightly less.

REDOX Process

The Bismuth Phosphate Process recovered only plutonium from the irradiated uranium feed. The REDOX process, based on hexone extraction, was developed to simplify the remote mechanical operations. This method was called the "REDOX process" because of its use of oxidation-reduction separations chemistry. It exploited the ability of methyl isobutyl ketone (hexone) to extract both plutonium and uranium (and also neptunium) from oxidizing solutions

(Hill and Cooper, 1958). This counterflow extraction process was carried out in stainless steel equipment and used column cascade extraction technology. The aqueous feed consisted primarily of molar nitric acid that contained the fission products and transuranic (TRU) elements as nitrate salts. Sodium dichromate oxidant was used to ensure that both the plutonium and uranium were in the hexavalent state (as uranyl and plutonyl ions). In a plant with enough stages, methylisobutylketone (known as hexone or MiBk) extracts these ions from the fission products virtually completely. In this process essentially all of the radioactive fission products and the excess oxidant and its reduction product were rejected to waste in the first process step, minimizing oxidation and radiolysis problems of the solvent and radiolysis of downstream reagents needed for plutonium isolation (Culler, 1956:201-211).

Recovery of plutonium from the organic solvent was achieved by contacting the organic stream with a dilute nitric acid aqueous phase that was heavily salted with aluminum nitrate and contained a moderately strong reducing agent (Culler, 1956:172-194). Ferrous ion was selected to reduce $PuO_2{}^{+2}$ to Pu^{+3}, but not $UO_2{}^{+2}$, which remained in the organic phase. The uranyl ion was stripped from the solvent with dilute nitric acid later in the process. Like the Bismuth Phosphate Process that exploited the differences in solubility of plutonium phosphate between Pu^{+4} and $PuO_2{}^{+2}$ oxidation states, the REDOX process utilized changes in the plutonium oxidation state to effect a separation from fission products and other actinides.

PUREX Process

Following the Manhattan project, many solvent systems were investigated for use in plutonium extraction (Culler, 1956:172-194). Much effort was concentrated on the alkyl phosphates after it was found that TBP had useful extraction characteristics for both plutonium and uranium recovery. None of the other candidate solvents at hand at that time had characteristics significantly better than TBP for plutonium and uranium extraction. An advantage of TBP was that it allowed use of 8 M nitric acid as the process salting agent, whereas most other extractant systems required aluminum, magnesium, or some other highly soluble nitrate in concentrated solution. TBP was also available in large quantities in a very pure state at a reasonable cost.

When diluted to 30% by volume with kerosene, TBP extracts Pu^{+4} and $UO_2{}^{+2}$ with distribution coefficients (K_d) of about 20 from 6 to 8 M nitric acid. It is not as flammable as hexone and is highly immiscible with nitric acid solutions. It is reasonably stable to radiolysis and hydrolysis during processing and can be purified from decomposition products by contacting it with an aqueous sodium carbonate solution. Because of its chemical process simplicity, TBP became the successor extractant to hexone for aqueous separation processes.

The PUREX process resembles the REDOX process in that essentially all fission products are rejected at the first stage of the extraction sequence. The feed is a much simpler solution of uranyl nitrate and the other elements in 6 to 8 M nitric acid, with a trace of nitrite present to stabilize Pu^{+4}. Only uranium (as $UO_2{}^{+2}$), plutonium (as Pu^{+4}), and neptunium (as $NpO_2{}^{+2}$) are extracted from the nitrate feed; other salting or special oxidizing agents are not used. The uranium and plutonium are decontaminated together, and the partitioning of plutonium from uranium is done by hydroxylamine reduction of Pu^{+4} to Pu^{+3}, which is then back-extracted into 6 M nitric acid while $UO_2{}^{+2}$ remains in the solvent phase. The decontaminated Pu^{+3} is removed from the organic stream, free of uranium, and the uranium fraction is recovered by back-extraction in very dilute nitric acid.

Because PUREX does not need process salting to work efficiently and both solvent and aqueous reagent streams can be reclaimed for recycle, the volumes of waste fluids are greatly reduced. The fission product fractions can be discharged in concentrated form to holding tanks for radioactive decay prior to final vitrification and disposal. The process system works very reliably and produces a plutonium product with a decontamination factor from fission products of greater than 10^7. The uranium fraction has a similar decontamination factor.

Nonaqueous Processes

Fluoride Volatility Processing

Uranium hexafluoride production processes were initially developed to produce feed for the gaseous diffusion process for uranium enrichment and became routine on the multiton per day scale. The final purification of the uranium from ore is usually by solvent extraction, and the recovered uranium oxide is reduced to the dioxide, hydrofluorinated with hot anhydrous hydrofluoride, and then fluorinated with fluorine from a nonaqueous electrolytic cell. When it was found that plutonium also formed a volatile hexafluoride with characteristics very similar to uranium hexafluoride, it became apparent that plutonium and uranium might easily be recovered from irradiated fuels by volatility processing. Few other elements form volatile fluorides. Exceptions among the fission products are molybdenum, technetium, ruthenium, and tellurium. Tellurium (as Te_2F_{10}) is the only long-lived fission product closely following UF_6 chemistry, while iodine (as IF_5) follows UF_6 chemistry to a lesser extent. However, both elements can be oxidized to higher oxidation states (TeF_6 and IF_7) that are much more volatile than UF_6 and PuF_6, and they can be separated from the uranium product by simple distillation methods.

A process was demonstrated for purifying plutonium residues by fluorinating PuO_2 in a fluidized bed reactor with fluorine at 400° C to form PuF_6. After purification the PuF_6 was decomposed to PuF_4 and F_2 in a thermal decomposition column (Hyman et al., 1956). This system worked satisfactorily but was not put into production. More recently, a development effort was conducted through pilot-plant testing to demonstrate the use of fluorine oxide, O_2F_2, to convert PuO_2 to PuF_6 at ambient temperatures in simple reactor systems. Isolation of plutonium or uranium from bulk impurities or fission products by volatility methods has been demonstrated to be a practical approach that could be scaled to industrial levels.

Fluoride volatility processing can also be used to recover neptunium (as NpF_6), but there are no known hexafluoride compounds of americium, curium, or any of the other actinide elements heavier than plutonium. Therefore, its proposed use as a recovery technique for actinide elements from molten fluoride salt systems appears limited to volatilization of uranium, neptunium, and plutonium among the actinides. An alternative isolation method would be needed for the other actinide elements.

Uranium and plutonium hexafluorides are powerful fluorinating agents that require use of nonreactive materials of construction. Monel, Inconel, and certain aluminum and copper alloys have been used successfully over the years in UF_6 process systems, but shaft seals and packing glands pose significant challenges. Fortunately, proposed process systems have few moving parts.

PuF_6 is susceptible to internal radiolysis, with a significant decomposition rate to PuF_4 and F_2. The reaction is reversible, and in a closed system there would be an equilibrium composition dependent on pressure and temperature. Therefore, means to refluorinate the PuF_4 residue must be built into any system handling PuF_6 in quantity. Neutron emission occurs from PuF_4 and PuF_6 by (α, n) reactions on fluorine, and neutron shielding for personnel must be included in the design of all plutonium fluoride systems.

Large-scale fluoride volatility plants for spent nuclear fuel reprocessing are certainly feasible, but none have been built in Western countries. A pilot plant utilizing fluoride volatility as a component of the overall plant system has reportedly been constructed in Dimitrograd, Russia, for the processing of fast-reactor demonstration fuels. Little information has been released on the utility of this prototype facility or on the economics of volatility processing under the conditions imposed by spent fuel.

The existing large-scale fluorination systems that are used in the preparation of pure UF_6 for the separation of the isotopes of uranium obtain very good separation from most elements. There have been difficulties with the separation of the very small amounts of molybdenum, technetium, neptunium, and plutonium that have been present in the feed derived from the recycle of reactor fuels. Technetium progresses very slowly through the diffusion cascades and is a constant problem once the system is contaminated with it. Tungsten and molybdenum hexafluoride move readily in the diffusion plant and can slightly contaminate the product.

Molten-Salt Processes

Two basic types of molten-salt systems have been used in separations: (1) the mixed fluoride salt fluid that was used as a coolant and a homogeneous fuel and blanket system in the Molten Salt Reactor Experiment, and (2) the simple chloride eutectic salts that are used as an ionic solvent in pyrochemical spent-fuel reprocessing systems that are intended for use with highly irradiated metallic reactor fuels. There are preliminary results on the reduction of oxide fuels with molten lithium and lithium chloride. Between 400 to 600° C temperatures, lithium metal in conjunction with lithium chloride reduces actinide oxides completely. The resulting actinide metals can then be processed as if they were irradiated metallic reactor fuels.

In the Molten Salt Reactor Experiment application, the molten fluoride salt consisting of BeF_2, ^7LiF, ThF_4, and UF_4 was used as the working fluid. Molten salt provided a fertile thorium blanket, a neutron multiplier, the fissile fuel, and the reactor coolant, as well as much of the reprocessing solvent. The molten salt served as a suitable solvent for the bred ^{233}Pa, which was extracted from the salt phase into liquid bismuth by reduction with a controlled concentration of lithium metal. The fission products were removed from the flowing salt loop by processing a small stream diverted from the main coolant loop with a higher concentration of ^{7}Li metal. The purified salt was returned to the coolant stream after processing. The uranium was isolated as the volatile hexafluoride after fluorination with hydrofluoride and fluorine. The iodine was removed as hydro-iodine gas. The fate of the noble metals in the reactor was not resolved; they probably remained in metallic form as a suspension in the salt.

One of the desirable features of using ionic molten inorganic salts and molten metals for fuel processing is that both phases are inherently resistant to radiation damage effects. The main advantage of pyrochemical processing is the ability to chemically separate "fresh" reactor fuels that have very high concentrations of fission products, with their associated high decay heat output and intense radiation emission. In fact, the decay heat may offer a significant advantage, since pyrochemical processing typically is performed at process temperatures of 500 to 800° C.

Current pyroprocesses typically utilize the chemical-free energy between the molten element and its lowest oxidation state as an ion in a molten-salt solvent in a molten-chloride salt phase. Pyrochemical extraction chemistry does not have the complications of hydrolysis and chemical instability of aqueous extraction chemistry. Much of the needed thermodynamic data is at hand or is easily derived. Key to the evolving technology is the use of lithium chloride as part of the salt mixtures and lithium metal as a reducing agent. Magnesium chloride, copper chloride, and cadmium chloride may be used as selective oxidizing agents.

The salt transport process (Steunenberg et al., 1969), a pyrochemical method for recovering actinide elements from spent fast-reactor fuels, is an example of a sophisticated pyrochemical system. In this process, plutonium and uranium are recovered from fission product residues and other spent reagents. The basis of the separation is the transport of actinide metals between two molten alloys of magnesium, one containing copper and the other containing zinc. The alloys are chemically connected by means of an ionic molten-salt solvent, which permits movement of ions between the alloys. The driving force of the process is the difference in the thermodynamic activity of plutonium and uranium in the two alloys.

At 800° C equilibrium is established rapidly, and in only a few stages plutonium can be separated from uranium with a decontamination factor of about 100, while the fission products are readily isolated from the actinide elements present. The noble element fission products are retained in the copper–magnesium donor alloy, and the reactive fission products (cesium, strontium and the rare earth isotopes) remain as ionic species in the transport salt phase. No external electrical potential is needed in this process. The separation is accomplished by oxidation of the plutonium by the chloride solvent salt at the copper–magnesium interface and reduction of the plutonium chloride by the magnesium alloy at the magnesium-zinc interface. Mg^{+2} and Pu^{+3} move in opposite directions through the salt bridge.

Processes of this type can be readily designed to effect separations between chemical families of elements and can also provide separations between individual members of chemically similar families of elements. This high-temperature REDOX separation technique has yet to be developed to its full potential. It appears to be well matched to reprocessing of metal fuels used in fast reactors, where the burn-up will be high and the decay cooling time is expected to be short. There is need for liquid–liquid contactors designed for use with these systems and for reflux systems for the ends of cascades when high separations are desired.

Electrochemical Separations

The electrochemical technique known as electrorefining was developed for actinide systems as a simple way to purify plutonium metal alloys proposed as fuel for the Los Alamos Molten Plutonium Reactor Experiment (LAMPRE). The basic separations technique has been adapted to the purification of plutonium from several elements including americium, and to prepare pure uranium and plutonium metals for both the weapons and the breeder-reactor fuels programs (Coops et al., 1983; Christensen and Mullins, 1983). Electrorefining is classified as a pyrochemical process in this instance, because it also uses a molten salt as an ion-transfer medium between an impure metal anode and the pure metal cathode (the collected product). The process somewhat resembles the salt transport process described above, but the driving potential is provided electrically and can be controlled to provide a satisfactory cathode product.

In its simplest form, the electrorefining technique is based on the sequential oxidation of the most chemically reactive elements from a molten pool of feed metal (the anode) with transport of the cations through an ionic molten salt to the cathode, where it is reduced to metal and deposited. If the most reactive species present in the anode is an undesirable impurity, the cell potential can be controlled so that ion remains unreduced in the salt phase. In this way, the cell potential can be used to establish a narrow voltage window that permits either a single element to be deposited at the cathode or, in the case of multiple cathodes, different elements to be deposited sequentially and selectively on separate cathodes. When used in the latter mode, the electrorefining process becomes a batch processing technique. Impurity elements with a more negative chemical-free energy than the desired product remain in the transport salt. Elements with more positive free energies remain in the anode pool, either as a solution with the feed or as a sludge when the selected element is depleted from the anode.

Argonne National Laboratory Program in Pyroprocessing as Related to Integral Fast Reactor and Light Water Reactor Fuel Recycling

Pyroprocessing, as proposed by Argonne National Laboratory (ANL), has the advantages of high density, compact size, and fast kinetics as a consequence of the use of liquid metals, the high concentrations of the elements in molten salts, the temperatures employed, and the generally adequate element-to-element separation factors in the electrochemistry of molten-salt systems. The technologies required are under development and demonstration at present.

Integral Fast Reactor (IFR) Pyroprocessing Separations

The pyrochemical separations required for the integral fast reactor (IFR) pyroprocessing have been demonstrated at the laboratory and bench-engineering levels. These are mainly molten salt electrochemical processes for the metal fuel dissolution and the deposition of the uranium metal onto a solid cathode, with the plutonium and minor TRUs reduced from the molten salt into a liquid cadmium cathode. The technology at ANL appears quite feasible with respect to the IFR process. In ANL's program, the spent IFR fuel elements would be maintained in storage for about a year prior to processing to permit short half-like products to decay and eliminate the need for cooling the molten-salt baths during the pyroprocessing steps. Waste gases would be condensed via a cryogenic system involving the argon atmosphere of the working enclosures. They are not considered further in this document; this problem has been addressed by ANL.

Since these are mainly elevated temperature processes, the pyrochemical separation processes are dominated by thermodynamic rather than kinetic considerations. This is in contrast to most ambient-temperature electrochemical processes.

A process flowsheet is attached as Figure D-1. In the ANL pyroprocess, the minor actinides (americium and curium) follow plutonium. Data provided by ANL demonstrate that, in the process employed, elements of atomic number higher than uranium have similar but different thermodynamic properties. Consequently, large separation factors are observed between uranium and the other TRUs, but the separation factors among the individual TRUs are much smaller. In the ANL pyroprocess, elements are separated in groups according to their inherent thermodynamic properties. The transuranics behave as one group, uranium behaves differently, most of the rare earths behave as another distinct group, and so on.

ANL is in the process of defining the various primary and secondary waste streams. The process for IFR pyroprocessing, as it is currently being developed, will result in two new main high-level waste (HLW) forms. The first of these will be a natural or artificial zeolite for the waste salt stream containing the strontium, cesium, and iodide salts and the divalent rare earths that do not reduce readily to the metals and hence are partitioned to the electrorefiner transport salt phase. The second will be a metal matrix of copper, or copper-aluminum, containing the noble metals, some of the rare earths, and perhaps the fuel element undissolved hulls. Both the zeolite and metal matrix waste would constitute new waste forms for the repository.

The strontium and cesium fission products, as well as iodide and some of the rare earths, are retained in the KCl/LiCl molten-salt waste stream and are selectively sorbed from the salt by contacting the molten salt with solid zeolites. After hot pressing, a monolithic mineralized material would result (sodalite), which may offer potential as a new waste form. The exact form of the zeolite waste, however, remains to be determined. Preliminary experimental data from work with fine powders of a selected zeolite indicate that the zeolite leach rates can be at least comparable to borosilicate glass. The general concept appears sound with the available data indicating technological feasibility.

Regarding separation factors, both chemical models and some laboratory experiments indicate that LLW streams can be partitioned to contain less than 100 nCi/g of TRUs. The uranium that is produced for use as blanket pins has only traces of plutonium remaining in it, and the plutonium/minor actinide fraction appears to contain only small amounts of uranium, so that the driver feed can be formulated as needed. This is not a problem in terms of the fuel for the IFR. Since liquid cadmium is employed in the process, some cadmium would find its way into a melt and zeolite waste streams; ANL is attempting to minimize this. The IFR process depends in part on the thermodynamic driving force generated by the presence of cadmium and the addition of cadmium chloride as an oxidizing agent. Cadmium retorting, recovery, and recycle are essential to the success of the process.

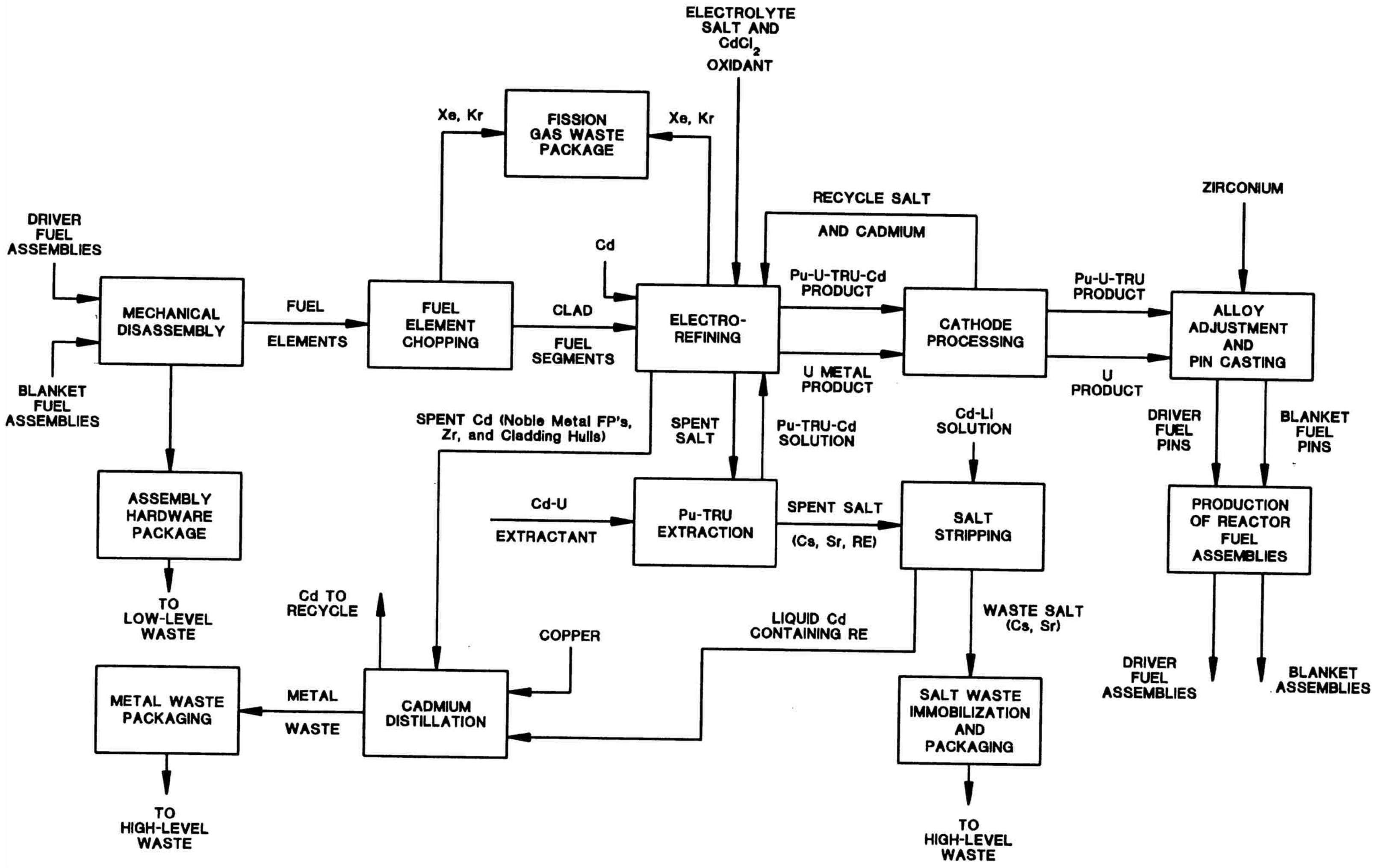


FIGURE D-1 IFR process flowsheet

Source: Argonne National Laboratory - Integral Fast Reactor Program, managed by the University of Chicago for the U.S. Department of Energy under Contract N. W-31-109-Eng-38.

Yttrium and the rare earths samarium and europium, which form divalent ions, and iodide remain in the molten salt and would therefore appear in the zeolite waste stream rather than the metal waste stream. These elements are not expected to present any problem with the cesium and strontium disposal schemes. The iodide might be recovered with a chlorination step.

The behavior of zirconium recycle for the driver alloy is not fully understood; additional thermodynamic data may be needed. However, this does not appear to be a critical process problem at this time. The IFR flow diagram indicates that some zirconium goes to HLW, and new salt must be added to the cycle.

A build-up of sodium, which arises from the dissolution of the clad sodium-bonded IFR fuel pieces, will ultimately raise the melting point of the molten salt eutectic. An alternative scheme for fuel treatment, in which the sodium and cesium would be distilled out of the fuel before electrorefining, is being considered. However, to maintain the proper salt composition, the molten salt electrolyte would need to be adjusted on occasion by discard and replacement or by a new processing step (i.e., preconditioned, lithium-loaded zeolite.

ANL is developing centrifugal contact reactors for use in cascades for "stripping"—reduction in this case—of the rare earths and residual TRUs from the molten salt stream with an alloy of lithium and uranium in cadmium. The bench scale efficacy of these processes has been demonstrated. This separation may require a multistage cascade with reflux using a continuous centrifuge contacting system.

Although waste volumes from the IFR recycle are currently an uncertainty, ANL estimates that they would be roughly comparable with those for direct disposal of unprocessed spent light-water reactor (LWR) fuel. However, the IFR wastes from the proposed pyroprocessing scheme would be essentially free from TRU elements (i.e., less than 1 ppm plutonium). The process is in an early stage of demonstration.

Light-Water Reactor (Lwr) Actinide Extraction/Separation Process

As a general comment, the pyroprocessing/separation aspects of this program are not as far along as the IFR pyroprocessing, since it has not been active for as long nor is it as well funded. This process requires that the oxide fuel employed in the processing be declad, and schemes for this are being investigated. Some initial work done in this regard at Oakridge National Laboratory (ORNL) appears to have had mechanical problems, and ANL itself has done a small amount of work on decladding (see discussion below).

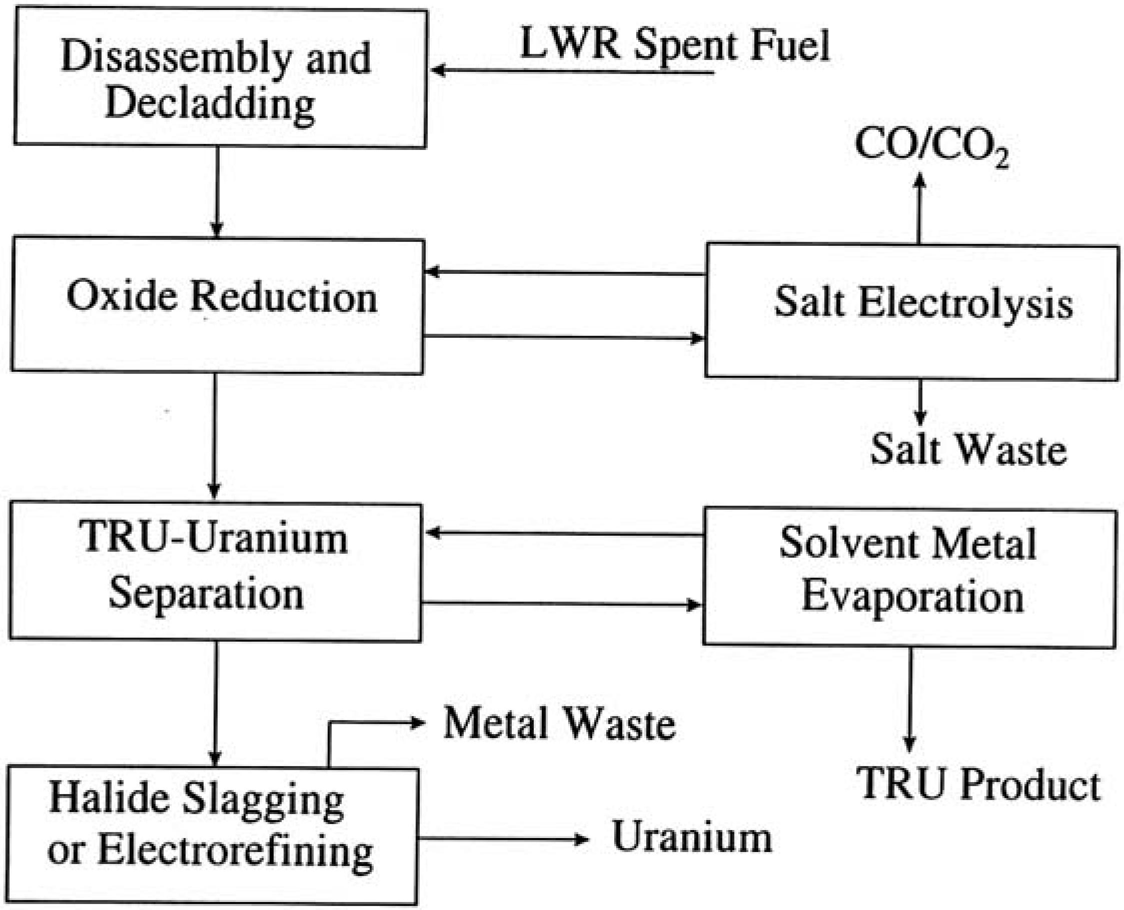
ANL is investigating electrolytic decladding using the Zircaloy hulls as an anode; this seems particularly interesting. The process could be considered as an electrowinning operation to remove the cladding and regenerate zirconium metal for recycle or disposal. The small amount of zirconium left with the oxide muds would not disturb the reprocessing in the IFR recycle. The fate of tin and aluminum in the system needs study. ANL looks favorably on this as a possible solution to the decladding problem; there are, however, some constraints as stated below.

It is important to realize that ANL's current program in LWR fuel reprocessing and the process to extract actinides from the LWR spent fuel are directly linked to the IFR pyrochemical process. The actinides removed from the LWR would feed into the IFR pyroprocessing scheme for further purification if needed and would be incorporated into fuel for the IFR in a burner mode. However, solely from the point of view of LWR reprocessing, the connection does not seem to be necessary. If this option were, however, to be considered for a stand-alone LWR processing cycle, significant modification of the two processes under active consideration would be required. In the LWR process as presently configured, the noble fission product metals and TRUs—and in one scheme, the rare earths—would be fed back into the IFR pyroprocess electrorefiner. A waste stream from the salt would be immobilized in zeolite before disposal; this is similar to the proposed IFR purification process discussed earlier.

Development efforts for the LWR actinide extraction processing are still in the early stages and are at low funding and manpower levels. There were three separate pyrochemical processes with variations under consideration for development when the STATS subcommittee members visited ANL in December 1991. As of September 1992, the number was reduced to two: a salt transport process and a zinc magnesium process. As of May 1993, there were still two processes under consideration, but the zinc magnesium process had been replaced by a lithium process and the salt transport process had been modified and subsequently has been abandoned. The elimination of the two processes previously under consideration, and modification of the salt transport process, eliminated the use of fluoride ions in the molten salt employed in the reduction step; fluorides caused degradation of the zeolites being considered as a waste form. Also, electrorefining has been added in both schemes under current consideration. The modified salt transport process had at first been chosen as the reference case, but more recently the lithium process has been favored, although further work is needed. As proposed, the lithium-based process offers lower operating temperatures that allows compatibility with stainless steel vessels and thus, considerable reduction in equipment costs.

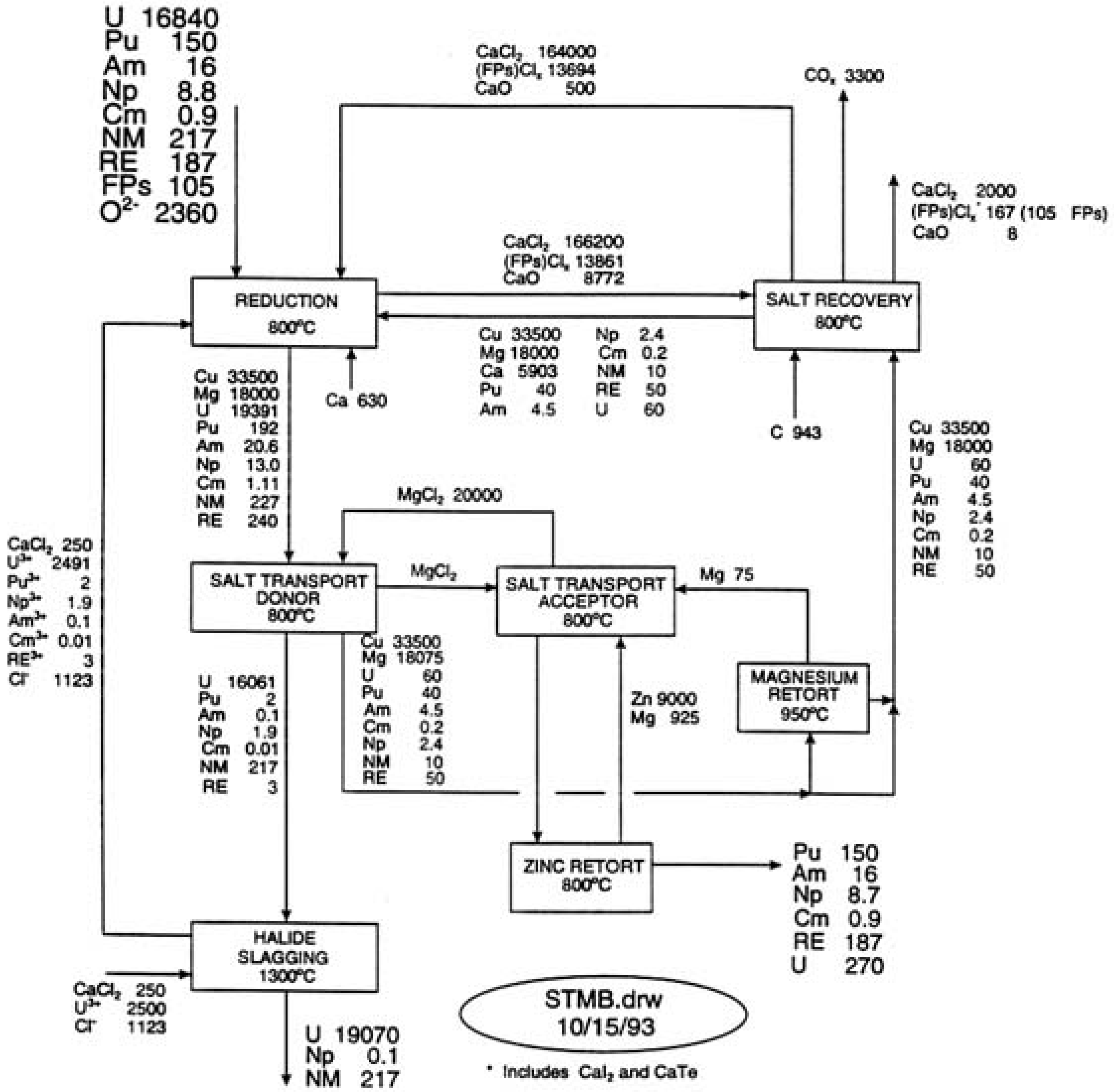
Figure D-2 is a generic scheme for both processes. Both involve (1) disassembly and decladding of the LWR spent-fuel elements, (2) dissolution and reduction of the oxide fuel in molten salt with a reducing metal phase present, (3) salt electrolysis to regenerate the reductant metal by oxide removal as carbon oxides with a consumable carbon anode, (4) TRU-uranium separation, (5) solvent metal retorting to recover the reducing metals and concentrate the recycle or waste streams, and (6) electrorefining to recover the pure uranium. Material balance flowsheets are included as Figures D-3 and D-4 for the salt transport and lithium processes, respectively. These must be taken as tentative, because the needed demonstration of the chemistry either has not been done or is not totally defined, as explained below.

In the salt transport process, the bulk of the uranium, TRUs, and rare earths would be in a copper-magnesium alloy following the initial oxide reduction step, which uses calcium metal as the active reducing agent. TRU and rare earth separation is effected using $MgCl_2$ as an oxidizing agent and transport medium to remove the TRUs from the copper-magnesium (donor) alloy, and the TRUs are reduced into a zinc-magnesium (acceptor) alloy from the $MgCl_2$ with recycled magnesium. The TRU-containing alloy is retorted to remove the zinc and magnesium, recover the TRUs, and recycle the solvent metals for TRU extraction. The bulk of the rare earths is expected to follow the TRUs, though some europium and samarium would appear in the salt waste steam as in the IFR cycle. The CaO, resulting from the initial LWR reduction step, is electrolyzed, CO_x is generated at the carbon anode and a magnesium-calcium alloy is



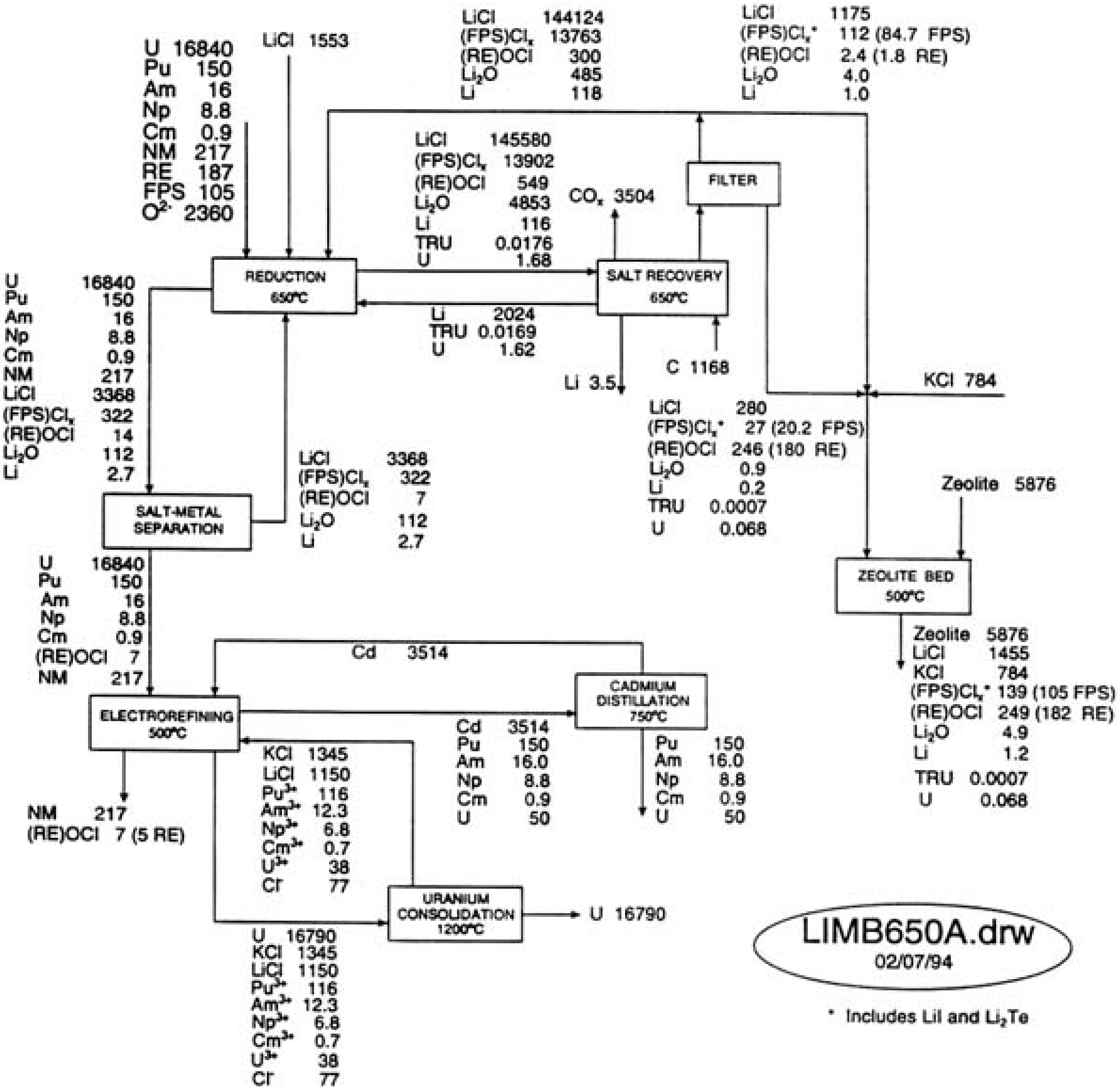
Source: Argonne National Laboratory - Integral Fast Reactor Program, managed by the University of Chicago for the U.S. Department of Energy under Contract No. W-31-109-Eng-38.

FIGURE D-2 Actinide recycle pyroprocess concept.



Source: Argonne National Laboratory - Integral Fast Reactor Program, managed by The University of Chicago for the U.S. Department of Energy under Contract W-31-109-Eng-38.

FIGURE D-3 Salt transport process material balance flowsheet (basis: 20 kg original LWR fuel, all masses in grams).



Source: Argonne National Laboratory - Integral Fast Reactor Program, managed by The University of Chicago for the U.S. Department of Energy under Contract W-31-109-Eng-38.

FIGURE D-4 Lithium process material balance flowsheet (basis: 20 kg original UO_2 LWR fuel, all masses in grams, LiCl reduction salt at 650° C).

produced. Following the salt transport removal of the TRUs and rare earths from the system, the uranium is electrorefined from the system to a dendritic form and melted in the cathode-processing step to separate occluded salts, which are distilled and recovered.

In the lithium process, the initial solvent salt is LiCl, and metallic lithium is employed to reduce the LWR oxide fuel. The LiCl, containing significant amounts of Li_2O, is then electrolyzed to lithium and CO_x using a carbon anode. Preliminary experiments indicate this is accomplished, with the lithium, uranium, and TRUs being reduced to the metals. Operations at temperatures of 650° C are reported to give the best results. The rare earths are expected to stay in the salt phase as oxychloride and to go into salt waste stream. However, the behavior of all rare earths in this process is not fully demonstrated. The uranium and TRUs are electrorefined from a KCl-LiCl salt bath. The TRUs are recovered in a cadmium cathode and the cadmium is retorted off, with the TRU fraction—and a small amount of uranium—serving as input to the IFR electrorefiner. Zircaloy, when used as an anode in the LiCl-oxide containing molten salt, appears to dissolve, but, in the presence of oxides, appears to form ZrO_2 as a precipitate. However, the initial experiments suggest this may be a route to hull decladding and zirconium disposal as ZrO_2.

Some rare-earth fission products would remain in the metal phase to be fed back into the IFR. In the process material balance flowsheet provided for the salt transport process, all of these products are estimated to feed back into the IFR, but see discussion above regarding europium and strontium following the cesium and strontium. In the lithium process, the rare earths are expected to form oxychlorides and follow the salt waste stream with the cesium and strontium fission products. The basic requirement is that the uranium to be recovered and stored can be adequately decontaminated and the volume of salt for discard remain at reasonable levels. However, more work needs to be done to make this a certainty. Regardless, this approach requires that the metal feed from the LWR process to the IFR be purified in the IFR electrorefiner before it is fabricated into fuel. The rare earths and noble metal fission products would follow the waste streams from the IFR process.

ANL has not yet published in the open literature process parameters documenting the proposed LWR process flowsheets, since the details of these proposed processes are "applied nuclear technology" and may not be made available to foreign interests unless specifically released by DOE. ANL has not yet demonstrated the complete LWR actinide extraction process on an engineering scale. However, an engineering-scale inert atmosphere drybox is under construction, in which one-tenth full-scale plant experiments, starting with the lithium process, would be carried out and experiments involving UO_2-simulated fuel were scheduled to be initiated during 1994.

The two alternative pyrochemical processes under consideration appear to be scientifically feasible. Most of the pyrochemical process steps have been demonstrated at laboratory scale only; as an example, an integral step of the salt transport process involves reduction of the spent LWR oxide fuel with metallic calcium, and the choice of a container suitable for carrying this out on pilot-plant scale, much less a commercial scale, has yet to be made. A major point in ANL's preference for the lithium process is that it operates at a lower temperature than the salt transport process, 640° C versus 800° C. This difference would permit use of stainless steel vessels for the lithium process. There are also distinct advantages in operations with uranium and iron when the temperature is well below the 725° C eutectic of the iron-uranium.

Although zeolites are proposed as a matrix for disposal of salt waste, the processing is still in such an early stage of development that is not now possible to identify the specific nature of the optimum zeolite and the products or waste forms from the actinide extraction processing. This scheme would probably also result in another new waste form for the metal waste as well; since the process to be developed has not been selected, it is not possible to further define the metal waste stream.

Although too preliminary to be at all specific, waste volume projections have been made by ANL personnel and suggest that the volumes would be roughly comparable with those for direct disposal of unprocessed spent LWR fuel.

Because of the very early state of process and equipment development, no attempt has been made to carry out a definitive cost estimate. It may be possible to make such an estimate after the engineering-scale process demonstration.

Questions have been raised concerning the fate of technetium during the pyroprocessing procedures. ANL contends that in the IFR and LWR work the technetium is sufficiently noble, and the molten salts involved maintained sufficiently anaerobic (with most steps involving elements to which technetium is noble) that technetium is expected to be found as the metal at all times. Based on a literature survey of the properties of technetium alloys and compounds, ANL concludes that technetium will most likely remain in the cadmium electrorefining solvent (anode) and be present in the form of intermetallic compounds in the IFR metal waste stream. It does not appear, however, that confirmatory work on this issue has been carried out at this time.

Reprocessing Experience In The United States And Abroad

At the present time, the international standard technology for aqueous spent-fuel reprocessing remains PUREX, but several alternative technologies are being considered: other solvents and new ion exchange techniques to extend the PUREX process, pyrochemical processes for reactors utilizing metal fuels, and fluoride-and chloride-based volatility processes for oxide fuels. If remote fuel fabrication is used, a decontamination factor of 10 to 100 is adequate for reactor fuel recycle, in contrast to the decontamination factor of 10^7 that is routinely required to permit plutonium processing by direct handling in simple glove boxes for weapons application. On the other hand, the defense applications have not emphasized the very high recoveries that are needed for the continuous recycle of the actinides through the proposed reactors system for the transmutation case. Losses to waste of less than one part per thousand per cycle are indicated as being needed. This objective can be obtained with the chemistries at hand, but not with the chemical engineering approach that has been adopted in the past, which used too few stages in the extraction systems to obtain the needed decontamination factors.

The separations facilities for the Savannah River Plant were designed to use PUREX solvent with mixer-settler contactors. The performance of PUREX exceeded expectations. It became the new "standard" in aqueous processing technology and was later installed at Hanford.

The Hanford PUREX facility utilized pulsed-column extractors and exhibited excellent performance. When operating on aluminum-clad uranium metal feed, the plant frequently exceeded its design rating of 8 tons of uranium metal per day. PUREX technology has been adopted in Europe for current nuclear processing plants, with ancillary equipment designed to fit applications. These applications include reprocessing Zircaloy-clad reactor fuels, recycle of plutonium as MOX for LWR commercial reactors, and prototype breeder fuels.

Experience at Various U.S. Sites

West Valley, New York

The first privately owned plant for reprocessing LWR fuels in the United States was built in the late 1960s and had a nominal capacity of 300 metric tons uranium (MTU)/yr. The plant, after obtaining an Atomic Energy Commission license, operated for several years with feed from the government's N-reactor at Hanford and from several LWRs in the United States. While the basic PUREX process worked well there, rapidly changing regulatory requirements and environmental rules forced the plant's shutdown in 1973. The new requirements made it less expensive to build a new plant than to modify the existing plant. Responsibility for the plant and the one full tank of HLW present at the site reverted to the state of New York and later to DOE. DOE is currently decommissioning the plant and fractionating the wastes to low-level concrete for permanent onsite storage, and to high-level borosilicate glass in stainless steel cylinders for shipment to the first federal HLW repository.

Morris, Illinois

A second 300 MTU/yr LWR reprocessing plant was constructed with private funds at Morris, Illinois in the early 1970s. This plant was designed to operate on a hybrid fluoride volatility and solvent extraction process, rather than the usual standard PUREX process. Before this process could be fully checked, it was realized that the plant would never have a sufficiently high operating factor to merit use; the plant had been built without buffer storage between its ten or so process steps, and no space had been provided to add hold-up tankage. Thus, all process steps in the plant had to run simultaneously or be entirely shut down. The process equipment was not sufficiently reliable to meet the former requirement, and no radioactive material was ever introduced into the separations plant. The fuel storage facilities have, however, received extensive use.

Barnwell, South Carolina

Between 1970 and 1975, the United States's first large (1,500 MTU/yr) reprocessing plant was constructed at Barnwell, South Carolina with private funds to reprocess spent fuel from some 60,000 MWe of U.S. utility-owned LWR reactors. This plant employed the basic PUREX process, using a centrifugal contactor for first-cycle extraction and pulsed columns for second-and third-cycle extraction.

The head-end of this plant was designed for remote operation and maintenance in a large hot cell. The downstream, lower-activity areas were designed to require no maintenance (all-welded construction, no moving parts) but were fitted with decontamination provisions for contact maintenance should any prove necessary.

By 1976, cold testing of the separations plant was underway, using natural uranium. A conversion plant had been constructed to convert the partially enriched uranium product to UF_6 for recycle to the gaseous diffusion plants for reenrichment and recycle to LWRs. The owners were awaiting Nuclear Regulatory Commission requirements for solidifying the plutonium nitrate product (conversion to PuO_2) and the aqueous waste (conversion to borosilicate glass) and were engaged in the final stages of licensing and resolution of issues in the Generic Environmental Impact Statement on Mixed Oxide (GESMO) fuels when U.S. government policies changed. In 1977, the Carter administration canceled the licensing and GESMO proceedings based on weapons nonproliferation concerns. Simultaneously, nuclear industry growth fell sharply, as did uranium prices, thus removing much of the economic incentive for LWR reprocessing. When government guarantees and assistance proved not to be forthcoming, the Barnwell plant was mothballed. Peripheral equipment vulnerable to corrosion or to becoming outdated was removed, but some 80% of the half-billion dollar investment remains intact in the form of seismically safe concrete structures and quality-assured stainless steel vessels, piping, and extraction cascades, etc.

Future Prospects

Existing federal waste policy legislation requires the U.S. government to assume responsibility for LWR spent fuel and its disposition. During the final phases of shutdown of the Barnwell plant, all interested U.S. companies stated that there would be no further private investment in reprocessing in the United States, and this attitude appears unchanged today. Thus, only the U.S. government could decide to reprocess LWR fuel in the United States in the future. Should the government elect to do so, the largely intact Barnwell plant could be made available for the purpose, according to its present owners.

Reprocessing does have many attendant advantages in the waste disposal process over direct disposal of spent fuel. Not only is some 96% of the spent fuel made available for potential beneficial use in fueling future reactors, but the residual waste is converted to a more compact, more chemically stable form.

This whole process may have uncovered the best long-range program for closing the tail-end of the nuclear fuel cycle—delayed reprocessing. All three of the LWR reprocessing plants constructed in the United States were designed to reprocess fuel 120 days after removal from the reactor. This was done at the West Valley plant. This calls for more stringent requirements about shielding, personal exposures, environmental releases in the event of accidents, and interim waste cooling requirements than would be necessary for the reprocessing of aged fuel. U.S.

industry has proven, by actually doing so, that it is capable of safely storing spent fuel at reactors for over 25 years. At this point, in fact, should the U.S. government decide to reprocess LWR fuel at the Barnwell plant, that plant could run for its projected lifetime without having to reprocess any fuel that had not been out-of-reactor for at least 25 years. This would not only provide much larger safety factors then those originally designed into that plant, it would also render moot the criticism made by some against designing the tail-end of the plant to allow the possibility for contact maintenance. After 25 years of cooling, the only fission products present in quantities hazardous to personnel performing such maintenance would be ^{137}Cs and ^{90}Sr. These are readily removed quantitatively and rapidly from equipment by washing with dilute nitric acid.

Experience Abroad

France

France is probably the most advanced nation in the world in the effective deployment of nuclear energy and in the resolution of fuel-cycle matters. Over 75% of that nation's electricity is currently derived from nuclear fission, and all of its spent fuel is scheduled for reprocessing. This is still less than the U.S. installed nuclear capacity. A government-owned company, COGEMA, is responsible for the nation's fuel reprocessing activities. For many decades, COGEMA has reprocessed spent LWR fuel at plants in La Hague on the Brittany coast. The older plants at La Hague have been upgraded and augmented so there are now two 800 MTU/yr plants being operated effectively by COGEMA at La Hague. These plants employ the PUREX process, have extensive underwater facilities for storing fuel awaiting reprocessing, and use remote operation and maintenance practices extensively. Fuel under contract for reprocessing there includes not only all spent fuel from French reactors but much fuel from Germany, Japan, and other European nations. These plants were designed for prompt reprocessing, but they are actually reprocessing fuel that has been cooled for several years due to logistic factors.

United Kingdom

The United Kingdom has for decades reprocessed spent fuel at facilities in northwestern England on the Irish Sea near Sellafield. In the early 1980s, design was initiated for construction of a new Thermal Oxide Reprocessing Plant (THORP) for reprocessing 1,200 MTU/yr of LWR fuels. Start-up of reprocessing operations began in 1994.

The THORP facility employs the basic PUREX process, has extensive prereprocessing storage for spent fuel, uses significant remote operations and maintenance practices, and was designed for prompt reprocessing.

Japan

Japan has also performed small-scale reprocessing of LWR fuels for many years at its Tokai-Mura plant. A large 1,200 MTU/yr LWR reprocessing plant is under construction on the coast in northern Japan by a Japanese consortium of nuclear industry organizations. This new plant has received considerable design assistance from France, and much of the technology employed in La Hague is being incorporated into the Japanese plant. This plant also employs the basic PUREX process. It is scheduled for start-up in 1998.

The Former Soviet Union

The former Soviet Union engaged in LWR reprocessing for many years, and some of the countries in that region are still significantly involved. A large fuel-reprocessing capability exists in several PUREX plants, and the suggestion arises periodically that international reprocessing services may be offered. Scientists in some countries of the former Soviet Union also continue to investigate advances in chemical separation of long-lived radionuclides, transmutation, and the vitrification process.

Other Nations

Several other nations have engaged in reprocessing of LWR fuels over the years. A 300 MTU/yr plant at Mol, Belgium, operated for many years under the auspices of Eurochemic, a consortium of 13 European nations. Germany has operated a small processing plant at Karlsruhe for decades. India has been engaged for years in small scale LWR reprocessing.

Future Separations Processes

This section surveys of a variety of separation technologies that have potential use in the transmutation processing systems or the nuclear wastes remediation program. Limitations of time and personnel determined the thoroughness of this survey and precluded an in-depth evaluation of every possible technology. In particular, promising separations and processing technologies from foreign countries were not systematically surveyed. For example, a series of Russian reports (published in 1991 by the Soviet Energy Technology Center of the Argonne National Laboratory) describe several novel and potentially quite useful technologies that should be considered in a more thorough review than the present one. This summary reflects the methods that came to the committee's attention during the period January 1992 through September 1992. The primary focus was on separation technologies that may be applicable to DOE wastes at Hanford, Savannah River, Rocky Flats, and other sites around the United States.

The evaluations are based on the experience of the members of the Subcommittee on Separations. None of the technologies are ready for full-scale use; some require more laboratory research (either basic or advanced) prior to pilot-plant testing, while others are ready for such testing now.

Possible Technologies for Actinide Separation

Solvent Extraction

Stereospecific Extractants. This category includes crown ethers (strontium extraction) and $Pu^{(IV)}$ complexants. Various crown ethers and related compounds have cavities of distinct shape and size, which can accommodate atoms of corresponding size. Such extractants (or the corresponding sorbents) can give very high selectivities. For processing of nuclear wastes, the role for such highly specific extractants should come late in the processing sequence, where it is desirable to remove or isolate a particular element. A crown ether is the current extractant of choice for the strontium extraction process developed at ANL for strontium isolation (Horwitz et al., 1990; Raymond et al., 1984).

Quite selective, stereo-specific agents for other problem substances such as cesium, technetium, iodine, americium, or plutonium would be of major value. Research at several laboratories on in vivo plutonium chelators has led to development of specific ligands for chelating $Pu^{(IV)}$ (Raymond et al., 1984). Similar ligands could possibly be applied to processing and removal of $Pu^{(IV)}$ from dilute aqueous waste streams. It might also be feasible to use them in high-salt waste streams.

Because of their stereo-specificity, it is often difficult to regenerate these stereo-specific reagents. Ease and completeness of regeneration are, therefore, vital criteria for agent selection. Along with rates of degradation, losses due to solubility and emulsification, and unit cost, the completeness of regeneration determines the rate at which the extractant is consumed, which can be an important cost factor for these relatively expensive substances. In order to reduce losses, it can be attractive to use specific functionalities attached to solid substrates, such as sorbents or membranes. The value that highly selective reagents would have in separations of waste components could be of considerable significance and justifies a major basic research effort.

Talspeak. The Talspeak process (Weaver and Kappelmann, 1964; Kolarik et al., 1972) is based on separation of lanthanides from trivalent actinides by extraction of the former into di(2-ethylhexyl) phosphoric acid (HDEHP) solution from aqueous phase of lactic and diethylene triamine pentaacetate (DTPA) acids at pH 2.5 to 3.0. The actinide transmutation of wastes (ATW) project has proposed a modification—the "reverse" Talspeak process—in which the trivalent actinide and lanthanide elements are extracted in an organic phase by HDEHP (Persson et al., 1986). The +3 metals are stripped into an aqueous phase of lactate and DTPA. Subsequently, 6 M nitric acid would be used to strip the lanthanides.

Relying as it does on the use of organic complexing agents and buffers, Talspeak may have some shortcomings in an intensely radioactive environment because of radiation damage.

This could cause problems in control of the process due to build-up of degradation products of unknown composition and effect.

Testing of the reverse Talspeak process at full radiation levels in pilot-plant operations would be needed before the process can be evaluated for full-scale processing.

Tramex in High Nitrate Solutions. High chloride systems with liquid anion exchangers such as trialkylamines or tetraalkylammonium salts have been used to extract and separate triand tetravalent actinides from most other fission products and the trivalent lanthanides. However, processing in chloride media is undesirable because of materials problems and neutron absorption problems. Studies have indicated that it may be possible to perform these separations from high nitrate salt solutions instead (Lloyd, 1963), but there are conflicting reports in the literature. This alternative use of nitrate solutions needs to be investigated further, as it could reduce the number of required processing steps, corrosion, etc.

Bidentate Extractants. This category includes carbamoyl phosphine oxides (TRU extraction, or TRUEX) and diamides and diphosphine dioxides.

TRUEX is an extraction process in which octyl(phenyl)-N,N-dibutylcarbamoylmethylphosphine oxide (CMPO)[2] is used to separate the TRU fraction from acidic LW solutions (Schulz and Horwitz, 1988).[3] The process has been demonstrated to work well at the laboratory scale, but pilot-plant demonstration has not been performed. An unanswered question about the TRUEX process that may limit its applicability involves separation of trivalent lanthanides from the trivalent transplutonium radionuclides.

A recent Russian report (Dzekun et al., 1992) supports use of diphenyl-dibutylcarbamoyl methyl phosphine oxide in polar inorganic solvents. Such solvents provide higher solubility without the addition of solubilizers. This extractant has also been used when sorbed on a solid porous support. Other bidentate organophosphorus extractants have also been studied. Among these, the diphosphine dioxides (Rosen and Nikolotova, 1991) and the diamides (Musikas and Hubert, 1987) also show good extraction and radiation-resistant properties but, like TRUEX, poor separation of trivalent actinides and lanthanides. In general, they are rather similar to the carbamoyl phosphorous extractants of the TRUEX process. Should use of the latter encounter difficulties in pilot-plant testing, these alternatives may be worth further evaluation.

The extracting agents in the TRUEX and stereospecific extractants (SREX) processes, a carbamoyl methyl phosphoryl derivative and a crown ether, respectively, have yet to be

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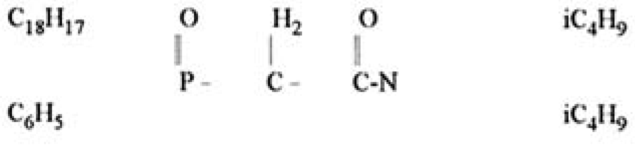
[2] The structure of CMPO is: CMPO-oxtyl(phenyl) = N,N'-diisobutylacarbanoyl-methylphosphine oxide.



[3] The reactions for TBP for TRUEX are: (1) Low acid: $AM(NO_3)_{3(aq)} + 3CMPO = Am(NO_3)(CMPO)_{3(o)}$; and (2) High acid: $Am(NO_3)_{4(aq)} + H_{aq} + 3CMPO_{(o)} = HAm(NO_3)_4(CMPO)_{3(o)}$

manufactured on a production scale, as will be needed if these processes are to be used for large-scale processing of nuclear wastes. Among the issues to be addressed are (1) effective precursors and processing methodology for manufacturing the extractants; (2) the purity achievable; (3) the nature and chemical effects of impurities; (4) radiolytic and chemical stability; (5) solubility in water or losses to emulsification, sorption onto suspended impurities, etc.; and (6) regenerability and degree of regeneration achievable. These factors could result in problems that would jeopardize use of the TRUEX and strontium extraction technologies and therefore they require prompt evaluation.

At present, TRUEX seems to be the most promising post-PUREX technology for TRU isolation from HLW. Pilot plant testing of TRUEX should proceed as rapidly as possible.

Soft Donor Complexants. Good separation factors have been reported between trivalent lanthanides and trivalent actinides for solvent extraction systems based on complexants with "soft" donor groups (e.g., nitrogen and sulfur). The complexants studied in France (Musikas, 1985) are based on amide functional groups. At Los Alamos, similarly good separations were achieved using sulfur based β-mik diketone extractants (Ensor et al., 1988). These systems are quite promising for trivalent cations. However, REDOX-based separations can separate the actinides thorium through americium more simply and, usually, more efficiently.

There seems little incentive or promise for developing soft donor ligands for the nontrivalent actinides compared with other more promising approaches involving stereo-specific ligands. However, they may have value in separations of transplutonium elements from fission product lanthanides.

Dicarbollide. Dicarbollide (bisdicarbollycobaltate), $(\pi\text{-}(3)\text{-}1,2\text{-}C_2\ B_9H_{11})_2Co^-)$, has been shown in Czech and Russian studies to have very high selectivity and efficiency for extraction of $Cs^{(+1)}$ and $Sr^{(+2)}$ from 2 to 3 M HNO_3 solutions (Rais and Selucky, 1992; Esimantovskii et al., 1992). The hexachloro and bromo derivatives have good chemical and radiation stability. However, dicarbollide, even in the presence of polyethyleneglycols, is less efficient for the extraction of trivalent actinides than for strontium and cesium but useful extractions have been demonstrated from 0.3 to 0.5 M HNO_3 solutions. Efficient extraction of cesium, strontium, barium, and americium from deacidified PUREX wastes has been achieved using a solution of 0.2 M dicarbollide with p-nonylphenol-nonaethyleneoxide (a polyethyleneglycol) with subsequent separation of lanthanum and americium species by their reextraction into nitric acid. The dicarbollides are available commercially. While they have desirable radiation-resistant properties, a disadvantage is that the waste would have to be made 0.3 to 3 M in HNO_3, depending on the process used, and the total volume of waste may be tripled.

There may be some value in the use of dicarbollides in comparison with use of crown ethers, cryptands, etc., but it is not evident at present that they could be employed satisfactorily on a large-scale basis. It seems unlikely that their use in the processing of waste that initially is highly alkaline should be considered.

Molten Salt. Researchers at Los Alamos National Laboratory are considering a molten salt system of $^7LiCl/BeCl_2$, which is based on the ORNL's research on a molten salt breeder reactor (Rosenthal et al., 1971). Liquid bismuth containing lithium metal would be used as an extractant in this system to remove the TRUs. The earlier studies of most of the reactor cycle at ORNL indicated in the molten-salt reactor project that this system may have a number of very promising features, but much more research and development is required for full assessment of its value in processing for the ATW system, etc.

Supercritical Fluid Extraction. Supercritical fluids (Li and Kiran, 1988) offer two potential advantages for separations. First, relatively small changes in temperature or pressure bring about substantial changes in fluid density, with resultant changes in solvation and reaction tendencies. Second, supercritical fluids can often be regenerated by a simple drop in pressure. Offsetting these advantages is the disadvantage of high pressure operation. No documented reports were found in which supercritical fluid extraction provides a separation for actinides, sodium salts, or fission products in a way that could be advantageous for processing of nuclear wastes, nor is there particular reason to expect such effects.

Ion Exchange And Adsorption

Organic Resins. A number of interesting new solid ion-exchange materials are in various stages of development. One of the most promising is Diphonix (Eichrom Industries, Inc), a substituted diphosphonic acid resin developed by a collaboration between ANL and the University of Tennessee. This resin, which is commercially available and is relatively inexpensive, is a very strong complexing agent and removes actinides from 10 M nitric acid solutions. It is reported to be very effective for removing a wide variety of toxic heavy metals (including lead, mercury, cadmium, zinc, nickel, cobalt, and chronium) from waste water and could be of use in removing various radionuclides from nuclear waste. Laboratory evaluation of such novel ion exchangers may result in useful separations for components of nuclear wastes.

Inorganic Exchangers. Inorganic ion exchangers such as titanium phosphate have been the subject of considerable research at a number of laboratories (Clearfield, 1982). The large capacity and high stability of such inorganic exchangers make them interesting candidates for study as ultimate waste forms for geologic storage. Some high-temperature zeolites may prove useful for removal of specific cations such as Cs^+. Recently, silicotitanates have been reported to have as much as 60 times the capacity of model zeolites for removing cesium from radioactive solutions of high salt content. The materials also remove strontium from such solutions and could be considered for use for the treatment of Hanford tank wastes. Sodium fluorophogopite mica, a clay, is reported to be superior to the zeolite clinoptilolite which has been used to remove strontium from nuclear waste (Paulus et al., 1992). This new clay mica removes cesium from solutions of high sodium content and can be prepared in bulk.

The initial research indicates that these systems have a reasonable possibility of application for specific separation and isolation of some fission products, but more study is needed for evaluation of this technology.

Adsorption. Adsorptive separations involve uptake of solutes onto solids with large specific surface areas, materials that act as solid solvent extractors, and materials with coordinating groups on the surface. Several of the approaches described in the subsection *Solvent Extraction* are adaptable to the adsorption mode, that is, functional groups such as ethers, ketones, ester, and crown ethers can be attached to surfaces of solids. In comparison with liquid-liquid extraction, solid adsorbents usually require the use of fixed or fluidized beds rather than simple countercurrent flow, although countercurrent flow has been utilized in pulsed towers and systems of tanks, but this may be offset by little or no residual solubility of the sorbent in the waste. A more subtle difference is that the interaction between the functional group and the solute occurs in an aqueous environment for an adsorbent, as opposed to an organic environment for extraction.

Researchers in Japan are investigating a process (McLafferty, 1992) wherein TRUs are adsorbed onto an unstated organic ligand fixed onto high-surface-area carbon fibers, tens of microns in diameter, enabling the TRUs to be recovered by combustion. (Licensing of such a process, however, is not easily permitted in the United States.) High surface area ferric, aluminum, and other hydroxides have also been used for clarification and recovery processes in the processing of nuclear wastes.

Adsorption processes may have limited capacity, but several systems have sufficient promise to warrant further research.

Membrane Processes

A number of different membrane processes can provide separations of interest for nuclear wastes. These include reverse osmosis, ultrafiltration, dialysis, facilitated transport, and electrodialysis. Most membranes used for these separations are polymeric and could, thereby, be vulnerable to radiolytic or chemical attack when used for the processing of nuclear wastes. Resistance to attack would be an important criterion for membrane selection. Ceramic membranes are also coming into use and may stand up better under the conditions required for processing nuclear wastes.

Ultrafiltration and Microfiltration. Ultrafiltration and microfiltration are similar to reverse osmosis, except that the terms are reserved for cases where the membrane retains high molecular weight solutes, colloids, or fine particles. Microfiltration could be used for dewatering of slurries or flowable sludges, where it would compete with conventional filtration or drying processes. High separation factors are available, and the method is already rather well developed.

Electrodialysis. Electrodialysis involves transport through stacks of anion and cation exchange membranes, across which there is an electric field. The process allows removal of salts from aqueous solution. Electrodialysis could be useful for the Hanford tank wastes if it were implemented in such a way as to remove the bulk salt components (e.g., sodium nitrate, nitrite, and carbonate) selectively, while leaving behind most or all of the various radioactive species. A considerable volume reduction of the waste would, thereby, be accomplished.

For selective removal of sodium nitrate and nitrite, it is desirable to have electrodialysis membranes that are selective for monovalent cations or anions, as opposed to multivalent ions. Such membranes, commercially available from companies in Japan (Tokuyama Soda and Asahi Glass), have been developed primarily for recovering NaCl selectively from sea water. The technique of manufacture is deposition of a very thin layer of anion-permeable material on top of a cation-permeable membrane, or vice versa. The anion exchanger serves to shield the membrane from multivalent cations. Another approach, with ordinary cation-or anion-exchange membranes, is to add a polyelectrolyte to the feed, which polarizes at the membrane and provides a similar shielding effect.

Research would be required to determine the resistance of candidate membranes to radiation, as well as to determine whether any components of the waste adsorb irreversibly on the membrane, to the detriment of the process.

Facilitated Transport. Facilitated transport membranes are impregnated with a chemical agent similar to the extractants used in solvent extraction processes. Regeneration of the agent occurs on the product side of the membrane. The separations that can be accomplished parallel those that are feasible by solvent extraction with the same or similar agents. The solvent extraction processes work well, however, and it does not appear that the use of facilitated-transport membranes is more advantageous, except in cases where it is very difficult to separate the phases from one another physically (e.g., because of emulsification).

Reverse Osmosis. Reverse osmosis is used for concentration of aqueous solutions, with water passing through the membrane under sufficient pressure. Evaporation is a well-established process that can readily accomplish the same separation. It appears that there are no competitive advantages of reverse osmosis, except where a highly volatile solute of importance evaporates along with water. In this case, the solute is retained by the membrane in reverse osmosis.

Dialysis. Dialysis is a process wherein solutes with low molecular weight pass through a membrane, while larger molecules are retained. It does not appear that dialysis is likely to enable separations of interest for nuclear waste processing.

Volatilization

Fluorides. Several systems propose fluorination to remove volatile fluorides, especially UF_6. A fluoride volatility process holds considerable promise for reduction of fuel reprocessing costs below those of the currently employed aqueous process. This has been confirmed through a survey of previous states of nonaqueous reprocessing routes by British Nuclear Fuels. Areas under small-scale investigation include (1) improved uranium-plutonium separation, (2) simpler UF_6 purification, (3) intensified fluorination processes, and (4) alternatives to isolation of plutonium as the hexafluoride. Work at LANL on plutonium recovery using O_2F_2 and KrF_2 is an example of the possibilities of using fluoride volatility. For the proposed ATW, a molten salt system of LiF/BeF_2 is under consideration, based on the earlier developments of the ORNL-MSBR program (Fitzpatrick et al., 1992). A problem involves the "noble" metals, which do not form stable fluorides and form deposits on the containers and piping. There is also concern over the problem of entrained solids in the gas phase.

β-Diketones. Some β-diketone ligands form relatively volatile compounds with trivalent lanthanides (<10 mm Hg at 200° C) so similar volatility can be expected for trivalent actinides. By comparison, analogous uranium and plutonium tetravalent and hexavalent compounds have much lower volatilities ($\leqq 10^{-3}$ mm Hg). For β-diketonates such as "fod" (6,6,7,7,8,8,8-heptafluoro-2,2-dimethyl-3,5-octanedione), separation factors of greater than or equal to 10^4 for uranium and plutonium from $Am^{(+3)}$ can be expected (Steinberg et al., 1982). Fod is stable to air and water at temperatures $\geq 100°$ C and can be recovered by extraction with an organic solvent, but its radiation stability is uncertain. Fod is only one example of the class of β-diketone ligands that may be applicable. Processing of large volumes of aqueous waste by extraction and volatilization of the β-diketonate complexes presents rather formidable engineering problems. Separations using volatile β-diketonate compounds would not seem practical for the great volumes of defense wastes.

Chlorides. For zirconium-clad spent fuel, the cladding can be removed by forming volatile $ZrCl_4$ using Cl_2 similar to the ORNL Zirflex process (McLaughlin, 1992). LANL proposes use of a plasma torch. It is unclear whether the separated zirconium would require geologic disposal, and, in addition, the degree of release of ^{14}C and ^{129}I in the process is uncertain. The advantage over mechanical or aqueous chemical decladding is not apparent, especially given the probable time and expense required to develop the technique.

Atomic Vapor Laser Isotope Separation

In 1991, a National Research Council committee studied the Atomic Vapor Laser Isotope Separation (AVLIS) system for several separations, including reprocessing of spent-fuel wastes. Its report stated (1991):

> With regard to the application of the AVLIS process to high-level waste management, several possibilities have been suggested, such as the separation of all the actinide elements by using isotope separation, specifically the AVLIS process, instead of chemical separation. The rationale is that the high

> decontamination factors required to reduce the actinide inventory of the waste to the point where the maximum actinide risk is proportional to the inventory may be more readily achievable by using isotope rather than elemental chemical separation techniques. However, given the uncertain benefits of actinide partitioning, it is unlikely that reductions to these levels would be worthwhile, even assuming that such an achievement was technically feasible. Moreover, the cost of accomplishing this via the AVLIS process, using a different set of lasers for each isotope individually, is likely to be prohibitive.
>
> Other suggested applications involve the separation of individual fission product isotopes once the actinides and the fission products have been chemically separated. For example, ^{137}Cs could be separated and stored on the surface, thus reducing the heat generation of the high-level waste consigned to a geologic depository. However, the separation of a high-gamma-emitting isotope such as ^{137}Cs by the AVLIS process would involve a radical redesign of the technology developed for uranium and plutonium, which requires contact maintenance of the collectors at frequent intervals.
>
> Alternatively, the radiocesium could be chemically separated from the high-level waste and permitted to decay on the surface for several hundred years, after which it could be mined to separate the long-lived ^{135}Cs isotope for subsequent disposal in a geologic repository. Again, even if this were technically feasible by AVLIS, it seems unlikely that the costs would justify the waste management benefits unless a decision has been made that reprocessing of nuclear fuel is justified. This, in turn, requires a demonstration that the benefits of reprocessing with regard to waste management and resource extension outweigh the economic costs and proliferation risks. Until there is such a demonstration, the use of isotope separation techniques such as AVLIS does not merit further consideration.

The report stated as a recommendation that "AVLIS should not be considered for applications to commercial nuclear waste disposal until fundamental issues relevant to reprocessing are resolved."

There are isotope separation processes that are suitable for high radiation fields. One is the thermal diffusion process that has been used for isotopes from tritium to uranium. It involves columns heated on the inside and cooled on the outside and there are few pumping demands. There are ion exchange and solvent extraction process that could be applied with the usual limitations of these procedures in radiation fields.

Precipitation

Carbonate. It has been reported (Yakovlev and Gorbenko-Gemanov, 1956) that coprecipitation of Am^{V} (96-99%) and Pu^{VI} (99%) occurs with K_4 $UO_2(CO_3)_3$. Subsequently, Np^{V}, Pu^{V}, and Am^{V} were isolated as $MAmO_2CO$ $_3$ where M = NH +/4 , Na^+, K^+, and Rb^+. Pu^{V}

has been shown to sorb rapidly to about 80-90% on $CaCO_3$ while Pu^{VI} sorption was much slower, allowing a kinetics based separation of AmO +/2 and AmO 2+/2 species at pH 8 using $CaCO_3$ (Kobashi and Choppin, 1988). Separation of AmO +/2 and AmO 2+/2 in alkali solutions can be effected with carbonates in basic solutions but the efficiency is not likely to be 100% in complex wastes.

Natural Agents

Siderophore (microbial). This is a field that deserves further research, as it could result in important separation and concentration. It may be possible to use siderophores (microbially produced chelating agents) to sequester species such as Pu^{4+} from the environment (Wildung et al., 1987). Such agents may also be a factor in unwanted transport of plutonium in the environment. Modified siderophores or siderophore subunits may be designed to selectively bind Pu^{4+} or other actinides. For example, octadentate derivatives of desferrioxamine B have been shown to react strongly with $Pu^{(+4)}$ to form complexes and are being studied for removal of plutonium from waste streams (Whisenhunt et al., 1993; Neu, 1993).

Jimson Weed. Los Alamos has reported that when radioactive sludges were contacted with jimson weed, the plutonium was removed from the sludge and bound to the cell walls by a protein of the weed. It is noted that the jimson weed cells can sorb the plutonium whether they are dead or alive (Kumar, 1992). It has been known for some time that such cells accumulate uranium from waste streams by hydrolytic sorption on the walls. However, in high radiation fields, the destruction of the biological material may be a limiting problem.

Chitin. An interesting possibility for use of biological material has been reported (New York Times, 1992). A derivative of chitin, in the form of porous beads, showed potential for removing heavy metals from groundwater. The beads are collected with a magnet and the sorbed heavy metal is stripped. The process is now only at the level of laboratory study.

Transport

Fused Salt. In the salt transport process proposed for the IFR system (Chang, 1992), LWR spent-fuel oxides, principally UO_2, are reduced to metal in a liquid (800°C) metal plus fused salt system by reacting with calcium to form CaO. Uranium precipitates in the metal phase, and about 15% of the neptunium coprecipitates with the uranium. The concentrations of plutonium and the other actinides are below their solubility limits in the copper-magnesium-calcium alloy, and they accumulate in the liquid metal phase. Alkali and alkaline earth elements, tellurium, europium, possibly samarium, and iodine fission products remain in the salt phase, while the rest of the rare earths and the noble metal fission products accumulate in the copper-magnesium-calcium alloy.

The metal phase left in the reduction vessel after removal of the reduction salt is processed by the salt ($MgCl_2$) transport step in which plutonium, neptunium, americium, and curium displace magnesium to form their respective chlorides. Subsequently, magnesium in a copper-magnesium alloy reduces the plutonium, neptunium, americium, curium, and lanthanides that dissolve in the acceptor alloy. Noble metal fission products distribute between the donor alloy and the acceptor alloy, and the bulk uranium remains in the donor alloy as a precipitate. Recovery of the TRU elements can be 99.9% with no significant waste from this salt transport step.

The copper-magnesium donor alloy with uranium can be recycled to accumulate uranium until the latter precipitates. The uranium product is melted in contact with the $CaCl_2$-based salt containing a chlorinating agent such as UCl_3. The halide slagging results in partitioning of the coprecipitated neptunium to the salt phase from which it is recovered. The plutonium-rich heavy metal product from retorting the acceptor alloy product can be charged directly to an electrorefiner.

The separate steps of this process have been tested on a laboratory scale, and pilot-plant testing is to begin in the near future.

Magnetic Separation. Separation methods based on differences in magnetic susceptibility have been explored on a laboratory scale for the separation of uranium and plutonium from certain particulate wastes (Avens et al., 1990; Hoegler and Bradshaw, 1989). In principle, the technique could be considered for the separation of any of the paramagnetic actinides from diamagnetic or nonmagnetic materials. For large-scale application, this does not appear to be a promising method at this time, but it could be useful for special wastes such as pyrochemical salts and incinerator ash.

Electromigration (electrophoresis). In separations by this technique, an electric field is applied, and substances are separated from one another by virtue of differences in mobilities of charged species through an appropriate medium. Electrophoresis has proven very difficult to scale-up because of the difficulty of dissipating ohmic heat or destabilization of flows by convection currents brought about by that heat release. No electrophoretic devices currently exist with capacities adequate for nuclear waste treatment.

Centrifugation. The use of liquid high-speed cascade centrifugation in partitioning of HLW components is being investigated at LANL (Bowman, 1992). A theoretical study of the separation in a LiF-BeF_2 molten solvent of the actinides from fission products has indicated some promise for this technique. Laboratory demonstration of the separations is needed before centrifugation can be evaluated for possible use in HLW partitioning.

Possibilities for Technetium Separation

^{99}Tc (half-life 2.1×10^5 yr) is produced in high yield in the fission process—some 25 kg/yr per LWR and about 2,000 kg in the Hanford tanks. Not only is it a high-yield fission product but it forms a water soluble anion, TcO -/4 , which can migrate in natural waters. The TcO -/4 ion is slowly immobilized even in the upper layers of many soils that can reduce its impact on the environment by a factor 10 to 100 (Bonodiette and Francis, 1979; Hoffman, 1982). Transmutation via thermal neutron capture to stable ruthenium has been proposed (Jarvinen et al., 1992). Prior to either transmutation or storage of the long-lived ^{99}Tc in a waste form suitable for long-term storage in a repository, the technetium must be separated from the waste. In the case of transmutation of technetium to stable ruthenium, procedures for the separation of ruthenium and technetium also need to be developed. A significant fraction (but less than 50%) of the ^{99}Tc is contained in the dissolver solids (hardly soluble fines). However, this makes its separation easier. These are routinely recovered by centrifugation. Minimization and recycling of waste streams must also be prime objectives. The following are some possible separation techniques that could be considered for these purposes and for decontamination.

Solvent Extraction And Ion Exchange

The CURE study (Westinghouse Hanford Co., 1990) proposed a separate tail-end solvent extraction using a primary amine and pH adjustment with formic acid to remove technetium from the waste stream. It further proposed to use an amine extraction or ion exchange separation of the technetium remaining in the uranium nitrate product.

A technetium separation system based on the use of the liquid anion exchanger Aliquat 336, which has higher stability in radiation fields than TBP, has been proposed (Jarvinen et al., 1992). Improvements such as the use of centrifugal contactors and pulsed columns are proposed and conditions for efficient extraction and stripping of the technetium (and palladium) are being investigated.

Ion exchange separations of cationic ruthenium species show promise for removal of ruthenium from technetium.

Ozonolysis

Ozonolysis can be used in conjunction with volatilization or other processes requiring oxidation and offers the possibility of greatly reducing the amount of wastes produced. This has been discussed (Abney et al., 1991) in conjunction with the accelerator transmutation of technetium to stable ruthenium. A scheme was proposed for continuously separating ruthenium from technetium by volatilization of RuO_4 with the final products being stable ruthenium metal and O_2 and any residual TcO_2 which can then be returned to the transmuter (Walker, 1992).

Volatilization

Volatilization of RuO_4 after ozonolysis was discussed earlier. Other volatile species such as the fluorides RuF_6 and TcF_6 can also be used in separation schemes in which the oxidation states are selectively achieved (Abney, 1991).

Electrodeposition

It may be possible to selectively reduce and deposit technetium in the presence of ruthenium due to the differences in their REDOX potentials. The separations of ruthenium and technetium are probably only relevant in the transmutation scenario. Technetium is reduced to the corrosion-resistant metal by electrodeposition as a possible disposal form. Technetium alloys with the other noble metals should be even more corrosion resistant.

Magnetic Separation

The possibility of magnetically separating TcO -/4 (diamagnetic) from reduced forms of ruthenium which are paramagnetic is under consideration. The technique has even been used on solid suspensions in aqueous media (Abney, 1991).

Engineering Challenges to Separations

Handling And Drying Of Sludges And Slurries

The Hanford single-shell tanks typically contain a layer of sludge under a liquid, with a salt cake on top of the liquid. It would be useful to process the sludge, and in any event it would be necessary to move it. Air-lift circulation has been proposed at Hanford for mixing the sludge with wash liquids. If this method presents problems in suspending the sludge, it would be valuable to explore other methods of mixing and resuspension. Another need would be to dewater the sludge. Radioactivity would hamper the use of conventional drying methods. Consequently, novel approaches, such as electroosmotic dewatering, may be applicable.

Evaporators

Evaporation is already in use at Hanford and can be expected to have a prominent role in processing techniques for nuclear wastes. Fouling of heat transfer surfaces can be expected to be a problem. Because of difficulty of maintenance, it would be important to identify or generate designs for which the fouling problem is avoided or minimized. Vacuum evaporation may be attractive, since lower temperatures can lessen the occurrence of unwanted reactions.

Various forms of evaporators should be assessed for reliable and maintenance-free operation. Given the nature of wastes, noncondensible gases can be expected to collect in evaporators, and means must be incorporated to purge them so as to avoid substantial lowering of heat-transfer coefficients.

Extractors

Centrifugal extractors are effective for reducing equipment volume and residence time. It would be important to locate or identify designs that provide virtually maintenance-free operation for extended periods while avoiding build-up of solids and phases of intermediate density.

Engineering Opportunities

Data from Hanford and ANL show that a potentially useful separation occurs among the three layers in storage tanks. Actinides and some fission products concentrate into the sludge, because the hydroxides are insoluble under alkaline conditions. The sodium salts (nitrate, nitrite, carbonate) compose the salt cake, which probably also incorporates strontium and cesium. Other fission products should partition among the layers. Sludge washing and salt dissolution is proposed as a first step in the processing of the Hanford tanks and should be effective for reduction of sludge volume through removal of nonradioactive, soluble bulk (e.g., sodium salts).

Sludge washing, soluble salt purification, and ion exchange could usefully be staged, through implementation of the classical "Shanks-tanks" approach (King, 1980), as used in extraction of instant coffee and other applications. In this scheme, piping is provided such that the position of any one tank within a sequence of tanks is advanced one position at a time, through appropriate opening and closing of valves. One stage of washing, recrystallization, or adsorption could take place for each cycle. This provides the equivalent of countercurrent flow, without actually removing the sludge, ion exchange resin, other solid, or water insoluble liquid from the tank until sludge washing is complete. This approach increases the recovery of salts from the sludge and minimizes the volume of wash water, and the technique might be used for any of the aqueous operations with liquids or solids in the facility.

An opportunity should also exist for recrystallizing the salt cake, in order to purify it as much as may be needed to produce LLW. The approach is to redissolve the cake in an aqueous phase and then recrystallize, probably in an evaporative crystallizer. Again, this can be staged, with supernatant liquids transported countercurrent to the salt cake and with recrystallization by heating and cooling at each stage (e.g., by the Shanks system).

Summary

None of the proposed separation methods going beyond the basic ion exchange and PUREX with its auxiliaries in known equipment has been tested sufficiently to be employed immediately. Some are potentially applicable to defense wastes, others to transmutation (partitioning), and some to conventional reactor fuel reprocessing. Of course, some are of possible value in several such areas. A summary of the committee's evaluation of the technologies reviewed is given in Table D-1. The priority ratings reflect a combination of probability of successful development of the technology and the time at which the technology would be useful. For example, TRUEX is rated high based on its likely success and the immediate value of such a technology in processing the defense wastes. By contrast, diphosphine oxides and diamides are given a medium priority, as they would serve the same purpose as TRUEX but have not been as fully evaluated. Talspeak and Tramex are given medium priority ratings as they are not needed at the present time, even though these technologies seem quite promising. Thus, it is important to assess the committee's ratings as reflecting a proposed strategy for developing a program of separations research and development in which the urgency of the need of the technology and its potential for successful development are both reflected.

TABLE D-1 Summary of Potential Separation Technologies

Technology	Utilization	Development Stage	Time of Need	Priority
Separation of Actinides, Cs, or Sr				
Stereo-specific Extractants Crown ethers, SREX	Defense wastes reprocessing	Ready for pilot-plant tests	As soon as possible	High
Pu complexants	Defense wastes partitioning	Basic lab research	Whenever developed	High/medium
Talspeak	Partitioning	Advanced lab studies	Next decade	High/medium
Tramex	Partitioning	Advanced lab studies	Next decade	High/medium
Bidentate Extractants Carbamoyl Phosphine Oxides (TRUEX)	Defense wastes partitioning reprocessing	Ready for pilot-plant tests	As soon as possible	High
Diamides and diphosphines oxides	Defense wastes partitioning reprocessing	Advanced lab studies	Whenever developed	High/medium
Molten Salt	Reprocessing	Ready for pilot-plant tests	Next decade	High
Soft Donor Complexants	Defense wastes partitioning	Basic lab research	Whenever developed	High/medium
Dicarbollide Complexation	Defense wastes partitioning reprocessing	Advanced lab studies	Whenever developed	Medium
Super Critical Fluid Chromatography	Defense wastes reprocessing	Basic lab research	Whenever developed	Low
Organic Resins	Defense wastes partitioning reprocessing	Advanced lab studies pilot plant	As soon as possible	High

Inorganic Exchangers	Defense wastes partitioning	Basic to advanced lab studies	As soon as possible	High
Adsorption	Defense wastes partitioning	Basic to advanced lab studies	As soon as possible	Medium
Ultrafiltration Microfiltration	Defense wastes	Advanced lab studies	Whenever developed	High
Electrolysis	Defense wastes	Basic lab research	Whenever developed	High/medium
Facilitated Transport	Defense wastes	Basic lab research	Whenever developed	Medium
Reverse Osmosis	Defense wastes	Basic lab research	Whenever developed	Low
Dialysis	Defense wastes	Basic lab research	Whenever developed	Low
Fluorides	Reprocessing	Advanced lab research	As soon as possible	High
β-Diketones	Reprocessing	Advanced lab research	Next decade	Medium
Chlorides	Reprocessing	Advanced lab research	Next decade	Low
Atomic Vapor	Partitioning reprocessing	Advanced lab research	Whenever developed	Low
Precipitation	Defense wastes partitioning	Basic lab research	As soon as possible	Medium
Siderophore (microbial)	Defense wastes	Basic lab research	Whenever developed	High/medium
Jimson Weed	Defense wastes	Basic lab research	Whenever developed	Medium
Chitin	Defense wastes	Basic lab research	Whenever developed	Medium

Fused Salt	Reprocessing	Ready for pilot-plant tests	Next decade	High
Magnetic Separation	Partitioning	Advanced lab research	Whenever developed	Medium
Electromigration, Electrophoresis	Partitioning	Advanced lab research	Whenever developed	Low
Centrifugation	Partitioning reprocessing	Basic lab research	Next decade	Low
<u>Separation of Technetium</u>				
Solvent Extraction, Ion Exchange	Defense wastes partitioning	Advanced lab research	Whenever developed	High/medium
Ozonolysis	Defense wastes partitioning	Advanced lab research	Whenever developed	High/medium
Volatilization	Defense wastes partitioning	Advanced lab research	Whenever developed	High/medium
Electrodeposition	Defense wastes partitioning	Advanced lab research	Whenever developed	Medium
Magnetic Separation	Defense wastes partitioning	Advanced lab research	Whenever developed	Medium/low
<u>Engineering</u>				
Handling and Drying of Sludges and Slurries	Defense wastes	Pilot-plant tests	Whenever developed	High
Evaporators	Defense wastes	Pilot-plant tests	Whenever developed	High
Extractors	Defense wastes	Pilot-plant tests	Whenever developed	High
Engineering Opportunities	Defense wastes	Pilot-plant tests	Whenever developed	High

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Appendix E—

Defense Wastes—Additional Information

Hanford Disposal Costs For High-Level Waste Canisters

Costs for disposal of the Hanford vitrified high-level waste canisters can be broken into two elements: canister production at the Hanford site and canister transport to and disposal at the repository. Both of these elements are considered in the brief analysis that follows.

Canister Preparation

A high-level waste (HLW) vitrification plant is planned to be built and operated at the Hanford site. The plant will process wastes from the Hanford HLW tanks after appropriate treatment to produce vitrified waste in canisters for shipment to the HLW repository. A Department of Energy (DOE) report describes three treatment options for waste disposal and estimates the number of canisters to be produced as a result of each option, assuming that "defense canisters" are used (DOE, 1993a). The options are

Option 1	sludge wash	38,000 canisters
Option 2	actinide separation	11,000 canisters
Option 3	extensive separations	2,000 canisters

The canister considered is a thin-wall (0.375 in.) advanced design with external dimensions of 2 ft. (diameter) by 10 ft. (length). The numbers of canisters are best estimates based on various assumptions about how much material can be removed from the tanks.

The current best estimate available on the capital cost of the Hanford waste vitrification plant is $1.7 billion (Reckman, 1993). The plant as currently designed has a capacity of 372 canisters per year and a design life of 40 years, yielding total plant production of 14,880 canisters if the design production rate is achieved. (European vitrification plants are not meeting design production rates due to melter design and material problems, coupled with melter replacement difficulties. As a result, British Nuclear Fuels has recently undertaken the design and construction of an additional process line for its windscale vitrification plant at Sellafield, U.K.)

Labor figures are not available for the operation of the Hanford plant. However, the Savannah River Site's vitrification plant has a staff of 1,150. Assuming a 1,100-person staff at Hanford, and an average staff wage, including fringe benefits of $45,000 per year, the total labor cost over the life of a plant is $2 billion.

Current limitations and the design status indicate that if option 1, sludge wash, is selected, it will be necessary to build three plants, either in parallel or in series, depending on the time DOE, with the support of Congress, is prepared to take to complete the vitrification process. To a first-order approximation, there does not appear to be any significant economy-of-scale resulting from building three plants, except that plant design cost would not be expected to be duplicated. Therefore, the cost of the sludge wash option and the actinide separation option would appear to be the same per canister, based on a total life-cycle cost per plant of approximately $4 billion and either one or three plants depending on the option.

The extensive separations process offers the prospect that less vitrification plant capacity would be required to produce the reduced number of canisters resulting from the use of that process. For chemical process plants, the plant size should decrease with the cube root of throughput, implying a rough scaling factor of 0.33. Applying that factor to the life-cycle costs for option 2 is probably optimistic, since it surely understates the engineering and operating staff costs. However, it seems to be an adequate approach to a first approximation. Application of the scaling factor to the life-cycle cost for option 2 yields a comparison cost of about $2.3 billion.

The production costs for each of the options, to a first-order approximation based on this simplistic analysis, are as follows:

Option 1	sludge wash	production cost, $12 billion	$316,000 per canister
Option 2	actinide separation	production cost, $4 billion	$364,000 per canister
Option 3	extensive separations	production cost, $1.8 billion	$1.80 million per canister

Canister Disposal

DOE has performed a series of studies on the cost to the government of defense waste transport and storage in the repository. Single-repository and two-repository cases have been considered. This analysis considers only the single-repository case. The most recent available information is DOE's "Preliminary Draft of Cost Estimates for the Hanford Tank Waste Remediation Options Study Cases" (DOE, 1993a). A number of other reports provide detail on the methods by which costs to the government for each option are considered (DOE 1987, 1990a, b, 1993a, b). Elements include costs for transportation, tunneling, borehole drilling, and emplacement. The analysis provides results that are almost totally dependent on the number of canisters to be emplaced. The disposition cost per canister for each of the three options is

Option 1	sludge wash	$185,000 per canister
Option 2	actinide separation	$196,000 per canister
Option 3	extensive separations	$189,000 per canister

Total per-canister disposal costs, combining the per-canister cost for production and disposal for the three options, are

Option 1 (base case)	$500,000 per canister
Option 2	$560,000 per canister
Option 3	$2.00 million per canister

Savannah River Site Plans For A Defense Waste Processing Facility

Members of the STATS Subcommittee on Separations visited the Savannah River Site in South Carolina in February 1992, and were presented with plans for, and the status of, conversion of existing defense wastes at the site into low-level saltstone for onsite storage and high-level glass in canisters ("glass logs") for eventual shipment to and storage in the first federal repository for HLW. In April 1992, the entire subcommittee was given an overview of these plans at one of the subcommittee meetings. The HLW would consist of about 7,000 canisters about 2 ft. in diameter and 10 ft. long, each with about 3,700 lb. of glass. This would use about 4,000 tons of the federal limitation of 70,000 Mg of heavy metal content on the first repository.

The Defense Waste Processing Facility (DWPF) at the Savannah River Site (SRS) in South Carolina has been under construction for about 10 years and is now nearly completed. The DWPF process appears to represent an acceptable solution to the disposition of defense wastes at SRS. Results from initial DWPF operations should provide important insights for the designers of the Hanford Tank Waste Remediation System.

DWPF Process Description

The following DWPF process description was supplied to the subcommittee by Westinghouse Savannah River Company and is believed to represent an up-to-date effective summary of the process[1] (Baxter, 1986; Baxter, 1988). A diagram of the process is shown in Figure E-1.

Waste Processing In Tank Farms

The SRS waste is currently stored onsite in carbon steel tanks and exists in three forms: sludge, salt solution, and saltcake (formed through concentration of salt solution [Maher et al., 1981]). The sludge, which makes up approximately 10% of the stored waste by volume, consists primarily of precipitates of the hydroxides of iron, aluminum, and manganese. The salt (saltcake and salt solution) is largely sodium nitrate, sodium aluminate, and sodium hydroxide. The sludge contains most of the radioactivity in the waste, including small amounts of actinides not recovered in the reprocessing plants and most of the fission products, except for ^{137}Cs. The salt fraction contains most (around 95%) of the ^{137}Cs.

The salt (saltcake and salt solution) is decontaminated for disposal as LLW by removing the radionuclides by precipitation and sorption. Dissolved saltcake and salt solutions are pumped to an existing waste tank in the tank farm for pretreatment. A solution of sodium tetraphenylborate is added to form an insoluble double salt with potassium and cesium. At the same time, a small amount of sodium titanate slurry is added to adsorb residual strontium and plutonium in the salt solution.

The precipitate slurry is concentrated by continuous filtration, washed to remove the soluble salt, and pumped to the DWPF. After further processing, the concentrated radionuclides from the salt solution are immobilized as borosilicate glass (see below). The decontaminated salt solution is blended with Portland cement, slag, and fly ash in a separate facility for disposal as LLW (saltstone process).

Sludge waste is also pretreated by alkaline washing in existing waste storage tanks. The object is to dissolve nonradioactive ingredients such as aluminum, so that they may be processed with the salt solution as LLW rather than treated as the more costly HLW glass. High-aluminum sludges are leached with strong caustic to dissolve about 75% of the hydrated aluminum compounds and reduce the volume of this type of sludge by about 50%. All sludges are washed with alkaline solutions to reduce the soluble salt content of the sludge slurry. Current plans are to accumulate about 2,500 m^3 of washed sludge slurry and then to use the sludge slurry as batch feed to the DWPF. Each batch of sludge would supply the DWPF for 2 to 3 years of operation.

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[1] Material in this section and its bibliography relies heavily on revision 1B of Westinghouse Savannah River Company report WSRC-SW4-6, part 2, dated 4/92.

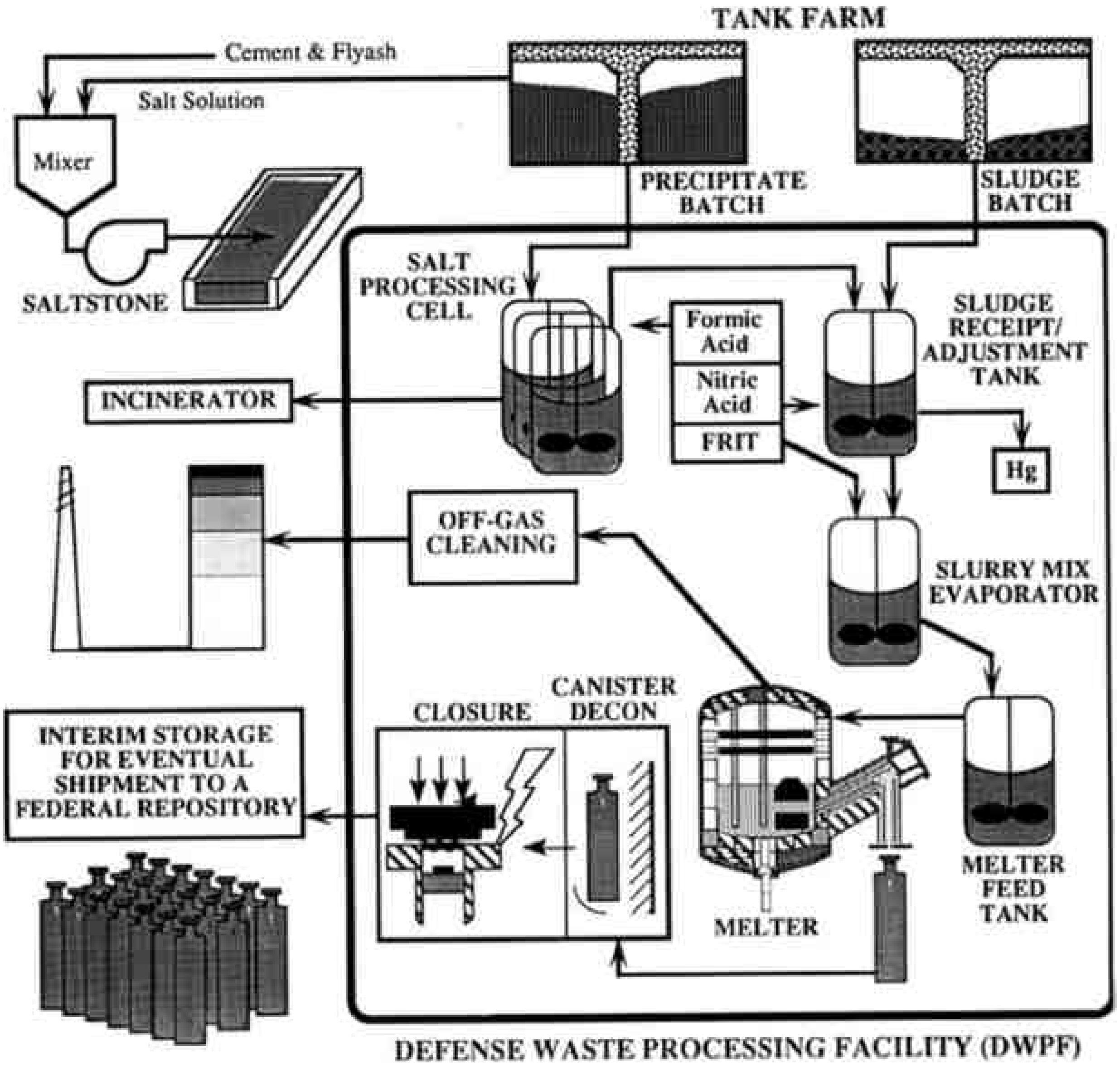


FIGURE E-1 Immobilization of Savannah River Site waste

Precipitate Processing

Within the vitrification building, tetraphenylborate precipitates are processed to remove organic material. About 90% of the phenyl groups of the tetraphenylborate salts are removed through reaction with formic acid and water (precipitate hydrolysis). The products of this reaction are an organic fraction (primarily benzene) and an aqueous product. The aqueous product contains the cesium as soluble formate salts. The aqueous product also contains some boric acid, formic acid, and about 10% of the original phenyl groups in water-soluble forms (such as phenol and phenylboric acid), along with the insoluble radionuclides—loaded titanates. The aqueous product is collected and fed to the sludge receipt and adjustment tank prior to vitrification.

Sludge Processing And Adjustment

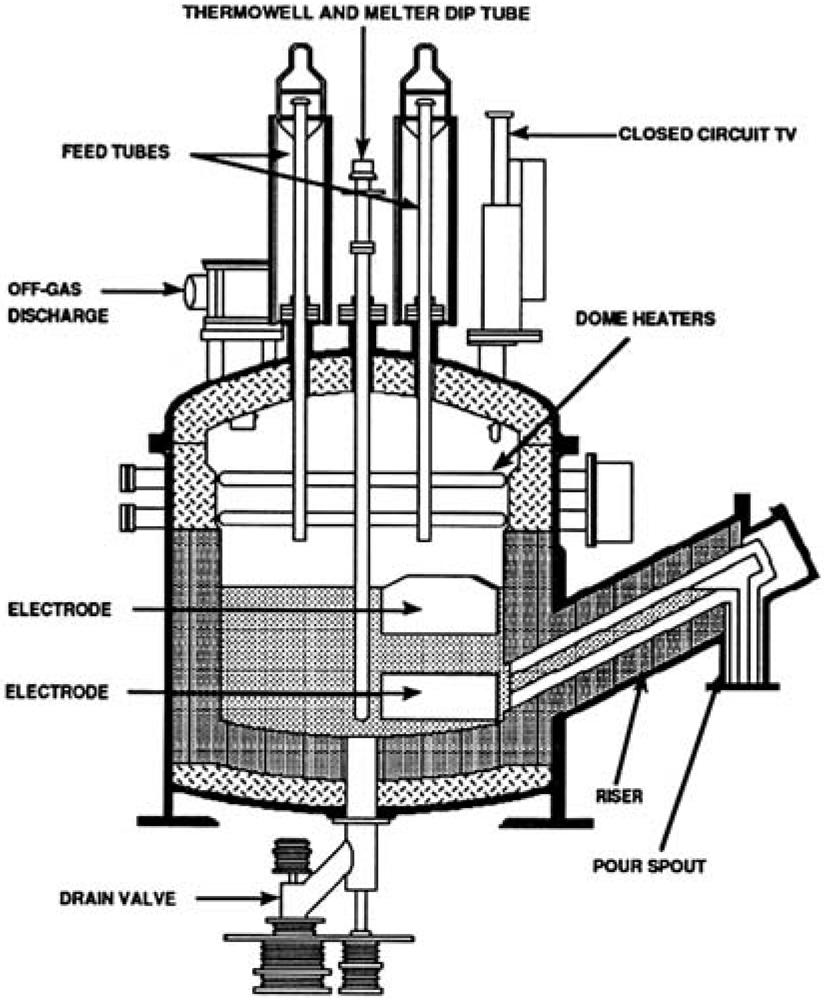
The washed sludge slurry and precipitate hydrolysis product are mixed together in the sludge receipt and adjustment tank. Formic acid is also added to the tank, which reduces any mercury ion present (a minor component of the sludge) to the elemental state. The formic acid also reduces the yield stress of the slurry and provides a reductant to the melt, which minimizes foaming. The mercury is steam distilled from the slurry and is eventually recovered in reusable, metallic form.

Waste Form Production

The slurry is transferred to the slurry mix evaporator where premelted borosilicate glass frit is added (Boersma, 1984; Weisman et al., 1988). Approximately two-thirds of the necessary frit is fed directly to the evaporator. The remaining one-third of the frit is first used for canister decontamination (frit blasting) and then is added to the evaporator. The frit-waste-formate slurry is concentrated to about 45 wt percent total solids by boiling. This mixture is transferred to the melter feed tank, which provides feed to the melter.

Vitrification of SRS waste is accomplished in a slurry-fed, joule-heated melter (Figure E-2). The feed slurry is introduced from the top of the melter. A crust, or cold cap, is formed on the surface of the melt pool as the water evaporates from the feed slurry and is removed via the off-gas system. Two pairs of diametrically opposed electrodes supply electric power directly to the melt. The nominal glass temperature beneath the cold cap is 1150° C, but varies throughout the melter. The cold cap melts from the bottom and forms the borosilicate waste glass matrix. For a nominal pour rate of 100 kg/h and a nominal glass melt weight of 6,500 kg in the melter, the average residence time in the melter is about 65 hours. The dome of the melter contains four pairs of resistance lid heaters that are used to provide the heat for start-up, as well as any supplemental heat required during glass production.

Glass is normally removed from the bottom of the DWPF melter through a riser and pour spout (Figure E-2). The canister, on the pour turntable, is connected to the melter by a bellows



MELTER CHARACTERISTICS

Weight of assembly filled with glass	73,000 kg
Total melter volume	5.5 m^3
Average glass volume	2.5 m^3
Glass height along sidewall	0.86 m
Glass height along centerline	0.95 m
Melter diameter	1.8 m
Pour spout inside diameter	5 cm
Riser angle	25° 30'

FIGURE E-2 Defense waste processing facility melter assembly, which seals the canister connection. Pouring is accomplished by drawing a vacuum on the spout relative to the melter. Canister filling is monitored by both a gamma-and a neutron-level detection system and by a weight system. When the desired level of fill in the canister is achieved, pouring is stopped by equalization of the pressure between the melter and pour spout. After pouring is stopped, venting of the canister to the off-gas system is continued for about 30 minutes to collect any volatile radionuclides (such as ^{137}Cs), which may be emitted from the surface of the glass. The canister is then rotated from beneath the pour spout. The canister is temporarily sealed with a tapered plug that shrink-seals as cooling continues, creating a leaktight seal of better than 2×10^{-4} cc-atm/s of helium.

Frit slurry blasting is used to remove contamination and metal oxides from the canister surface. Cleaning is performed by rotating the canister in an enclosed chamber and using air-injected wet glass-frit blasting on all exposed surfaces. The used frit slurry is sent to the slurry-mix evaporator for melter feed preparation.

Finally, the canister is sealed by welding a plug with a 12.7-cm diameter into the canister nozzle. After decontamination and drying, the temporary seal is pushed down in the canister neck, exposing clean metal for the permanent plug weld. The joint is clamped tightly and made as a resistively heated diffusion bond under pressure by a high amperage electric current. The weld produced is sufficiently leaktight based on experimental evidence, to approximately 1×10^{-9} cc-atm/s of helium and of comparable strength to the base metal to meet the required specification.

Interim Storage

The filled, decontaminated, and sealed canisters are moved by a shielded transport vehicle and stored in the glass waste storage building. Air is forced across the canisters' surfaces for cooling and to prevent overheating of the building. Although there should be no contamination in the building, the discharged air is passed through high-efficiency particulate filters so that any potential contamination is retained by the circulation system filters.

Ultimate Storage

The canisters are destined to be eventually shipped by rail to the first federal HLW repository. (Depending on when the repository becomes available, additional interim storage may be required at SRS.) Considerable effort has been spent over the past 5 years on tests to validate this waste form with the Nuclear Regulatory Commission for acceptance at the repository. It is anticipated that this waste will be suitable for acceptance by the agency for ultimate disposal in the first repository.

Comments on the DWPF Process

One problem with the DWPF process that is recognized by SRS personnel is the choice of tetraphenylborate to precipitate ^{137}Cs from supernatant solutions. This choice leads to many different organic compounds arising from thermal degradation. Some, such as benzene, have to be recovered and decontaminated as by-products. Still others have caused plugging problems in pilot-plant equipment. The reason that adsorption of ^{137}Cs on an inorganic zeolite—as was successfully done at West Valley, New York—was not selected at SRS is the resultant significant increase in the number of high-level canisters due to the volume of this zeolite. (This was not a significant consideration at West Valley, because there is only one tank of HLW to dispose of at that site.) However, the added cost burden that would result from zeolite absorption may be less than the costs of dealing with degradation products of tetraphenylborate at SRS. For future plants, it is possible that some alternative option would be superior to either the SRS or West Valley methods of ^{137}Cs collection.

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Appendix F

Transmutation Concepts

Advanced Liquid-Metal Reactor

Introduction

Liquid-metal reactor technology has been under development for over 40 years in the United States and several other nations. Its principal potential application would be as an advanced nuclear power reactor, capable of breeding new fuel from natural or depleted uranium. It has generally been assumed that the advanced liquid metal reactor (ALMR) will be needed in some future era when low-cost resources of uranium are depleted, so a breeder could avoid the rising fuel costs of light-water reactors. Although natural uranium is abundant and readily available on the world market, some nations, such as France, the United Kingdom, and Japan, have stated their intent to develop breeders to reduce their dependence on imports of natural uranium.

Although some of the early fast-spectrum reactors ran into serious problems, the more recent experience in the United States presents a very positive picture of fast-reactor technology. The performance of the EBR-II, in operation regularly since the early 1960s, and the 400 MWt (megawatt thermal) Fast Flux Test Facility, commissioned in 1979, have demonstrated that a medium-size liquid-metal-cooled fast reactor can be highly reliable.

The fast spectrum that makes an ALMR an efficient producer of fissile plutonium can also be used to transmute plutonium and minor actinides as an aid to waste disposal. Studies of fast reactors for transmutation were carried out in the late 1970s and early 1980s. In the late 1980s, the Department of Energy (DOE) project to develop the ALMR as an advanced power reactor focused also on applying the ALMR as a transuranic burner. France and Japan have shown similar interest. In the United States, this work has been conducted by the Argonne National Laboratory (ANL) and General Electric (GE), under DOE funding. For this application, core configurations that would operate at breeding ratios varying from 1.25 to 0.22 have been studied.[1] The reactor technology for transuranic transmutation is similar to, but more demanding than, that for the more familiar breeder application. Consequently, there is a large body of experience to aid the development of the ALMR as a burner of transuranic waste.

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[1] Breeding ratio is generally defined as the ratio of new fissile atoms produced to fissile atoms consumed. The ALMR project arbitrarily defines breeding ratio as the ratio of the number of atoms of new ^{239}Pu and ^{241}Pu formed to the number of atoms of ^{239}Pu and ^{241}Pu consumed by neutron absorption. The definition is arbitrary, because all atoms of plutonium, as well as the minor actinides, fission in a fast spectrum. With this definition, an ALMR core containing no ^{238}U could still have a finite breeding ratio because of neutron absorption in ^{238}Pu and ^{240}Pu. For an ALMR with no ^{238}U ANL estimates a breeding ratio of 0.22.

In this section, the ALMR work of ANL and GE is summarized, including the ANL proposal for an integral fast reactor (IFR), an ALMR with integrated fuel reprocessing, for power generation and waste transmutation.

Reference ALMR Designs

GE has the responsibility for reactor design for the ALMR project. GE's power reactor, innovative, small module (PRISM) concept is a modular ALMR generating 1,395 MWe (megawatts electric), comprising nine reactor modules arranged in three power blocks of 465 MWe each. As shown in Figure F-1, each power block in turn comprises three compact, sodium-cooled, pool-type fast reactor modules of 471 MWt with its own steam generator that is heated by secondary sodium piped from the intermediate heat exchangers in the reactor module. The design goals emphasize passive shutdown heat removal for loss-of-cooling accidents and passive reactivity reduction to a safe, stable state for undercooling or overpower events with failure-to-scram. The current PRISM-type ALMR uses a metal-alloy fuel that is being developed concurrently by ANL.

An ALMR can be designed as a breeder, such that the rate of production of new fissile material (plutonium and other transuranics) by neutron absorption in uranium equals or exceeds the rate of destruction of fissile material by fission and neutron capture. Such a breeder could be started in one of four ways: (1) with transuranic elements (TRUs) recovered from reprocessing spent light-water reactor (LWR) fuel, (2) with 25-35% enriched uranium, (3) with surplus military plutonium made available from the disarmament program, or (4) with excess plutonium produced by breeders of earlier generations. By reducing the amount of neutron absorption in uranium in the core and blankets, an ALMR can be designed to operate at a breeding ratio less than unity. This would require a continuous make-up of TRUs (e.g., from spent LWR fuel) as well as the start-up inventory, thereby increasing the mass rate of consumption of TRUs recovered from spent LWR fuel.

Two core configurations for the PRISM-breeder application and proposed TRU burner are shown in Figure F-2. The "actinide burner" core contains no radial and internal blanket assemblies to reduce the generation of new ^{239}Pu by neutron absorption in ^{238}U, resulting in a lower breeding ratio. The amount of TRU start-up inventory required depends somewhat on the breeding ratio at which an ALMR is operating. For a 1.4 GWe PRISM-type ALMR, the TRUs for a start-up core plus two reloads ranges from 27.2 Mg for a breeding ratio of 1.11 to about 14.6 Mg for a breeding ratio of 0.65.

ALMR designs with a wide variation of breeding ratios, over the range of 1.25 to 0.22, have been considered as part of the overall ALMR design study. Variations (Thompson et al., 1991) of the reference ALMR breeder design involve elimination of external blankets containing fertile material to reduce the conversion of uranium to TRUs. To increase neutron leakage, and thereby to reduce fissile breeding in the core, these burner designs require shortening the core height, with minor adjustment in fuel composition but without changes in fuel-rod diameter or in the number of fuel rods. This approach to enhance TRU depletion is based on the key constraint that the diameter of the reactor vessel remain constant, and it is obviously limited by

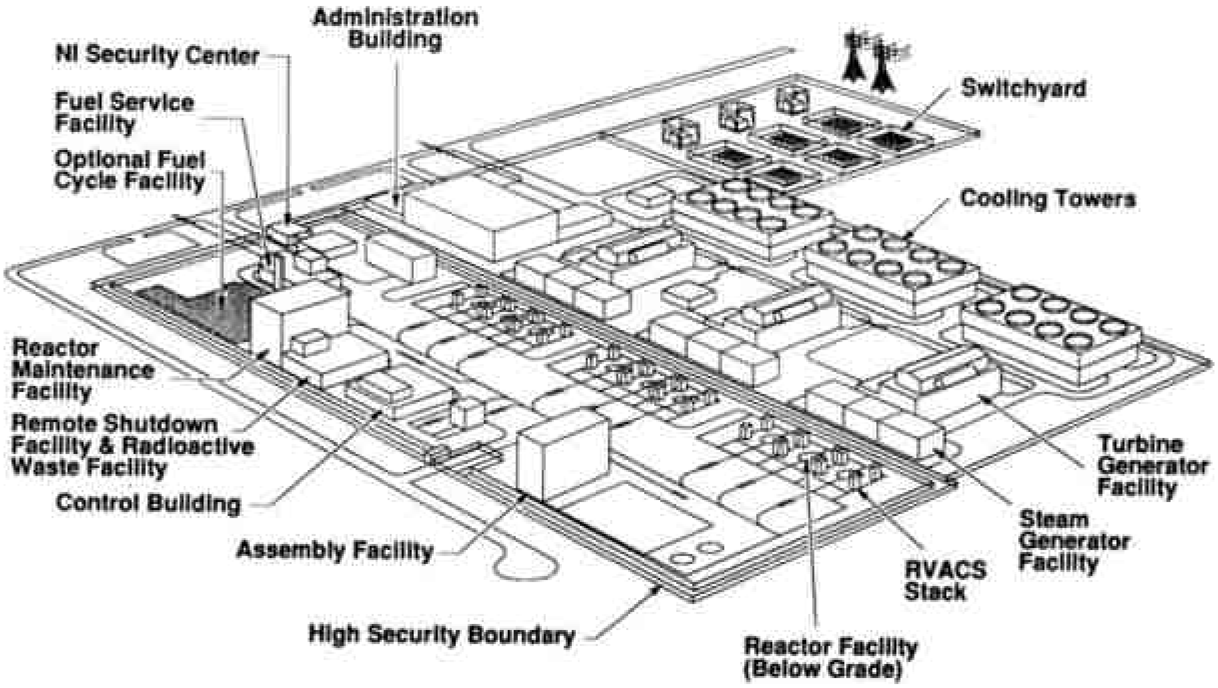
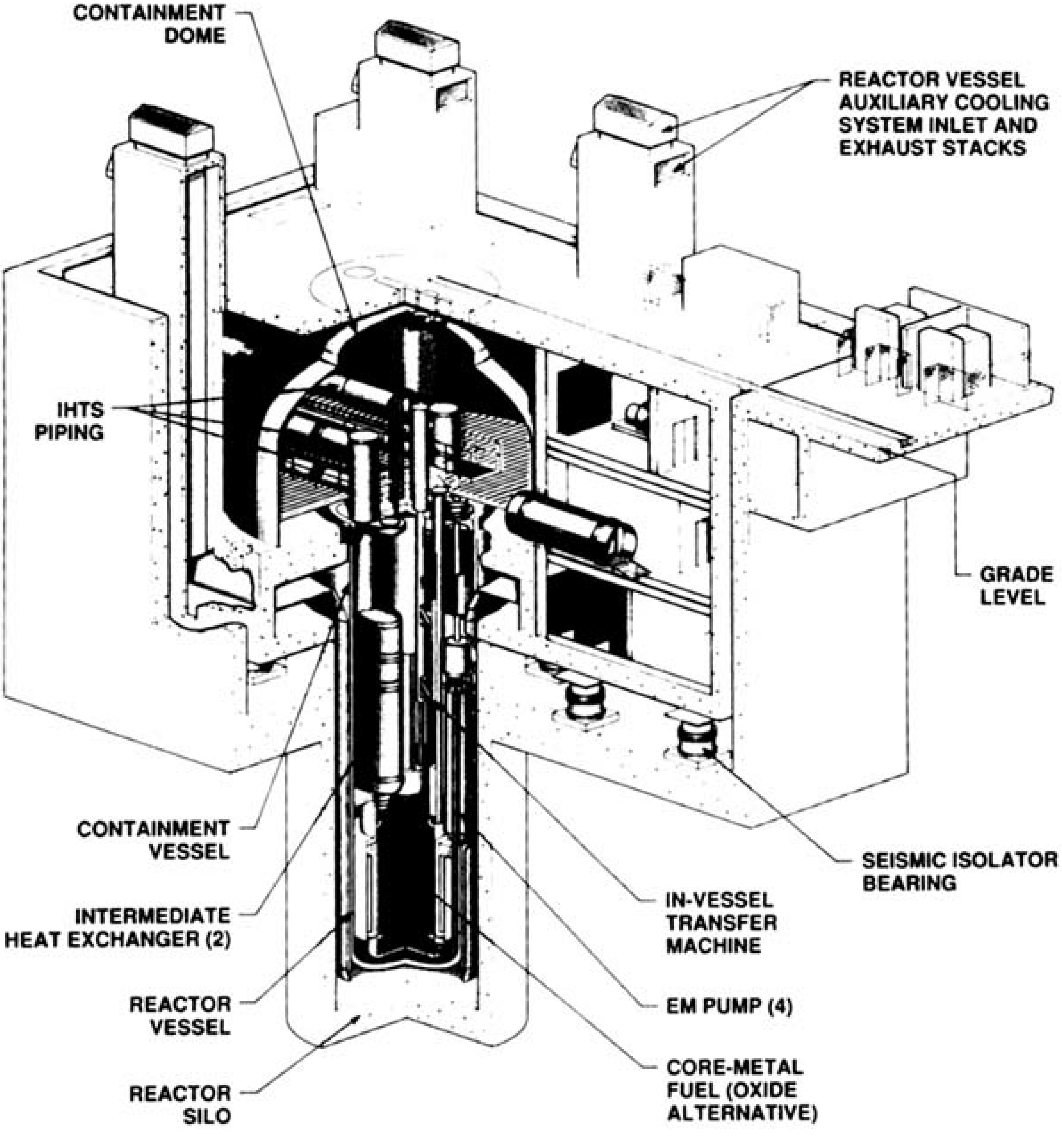
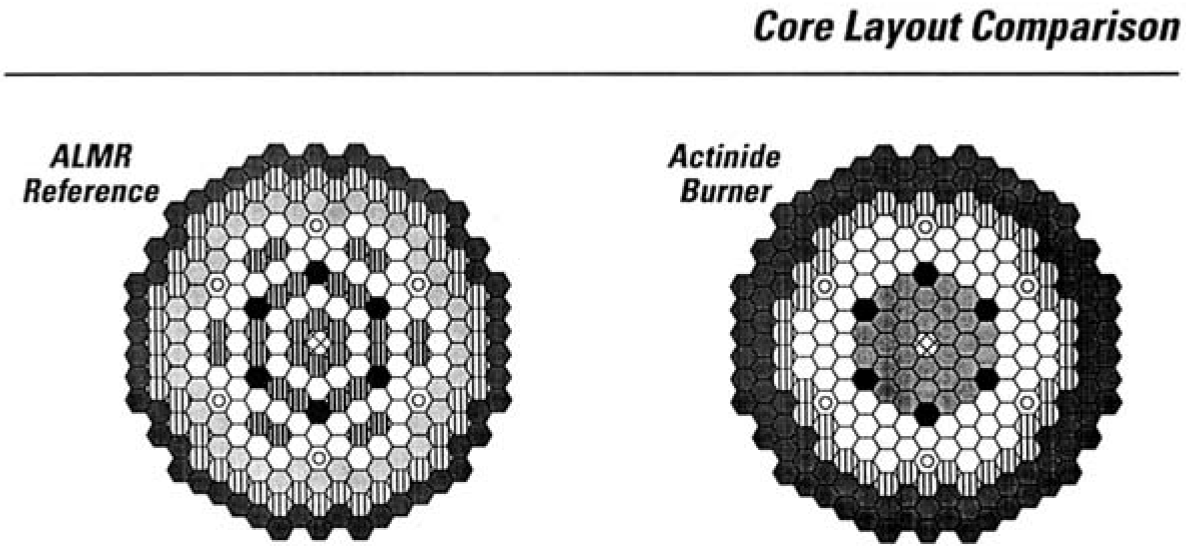


FIGURE F-1 ALMR power plant and ALMR reactor facility.
SOURCE: Quinn and Thompson (1992).

ALMR Reactor Facility





⬢	SHIELD	54
▥	REFLECTOR	48
◎	GAS EXPANSION MODULE	6
⬡	RADIAL BLANKET	42
⬡	DRIVER FUEL	66
▥	INTERNAL BLANKET	30
⬢	CONTROL	6
⊗	ULTIMATE SHUTDOWN	1
	TOTAL:	253

⬢	SHIELD	102
▥	REFLECTOR	42
◎	GAS EXPANSION MODULE	6
⬡	DRIVER FUEL (HIGH ENRICH)	66
⬡	DRIVER FUEL (LOW ENRICH)	30
⬢	CONTROL	6
⊗	ULTIMATE SHUTDOWN	1
	TOTAL:	253

FIGURE F-2 Core layout comparison.

the maximum allowable power density or by the linear heat generation rate of the fuel rods, so that safety characteristics are not compromised.

With reduced fissile breeding and reduced heavy metal inventory, the burner designs also result in increased reactivity swing over a fuel cycle. This then requires larger control rod worths and hence entails potentially larger positive reactivity insertions and degraded performance in transient overpower events. As the breeding ratio decreases, there is less reactivity insertion resulting from sodium voiding in a power excursion. However, with decreasing breeding ratio, less negative reactivity is available from Doppler broadening of the neutron absorption resonances that occurs when the fuel is heated in a power excursion.

Based on these considerations, GE concludes that a TRU burner with a breeding ratio of 0.60 and a core height of 0.76 m (30 in.) is the lowest possible breeding ratio configuration that would have acceptable safety features.

Designs with breeding ratios varying from 0.22 to 1.25 appear in the ALMR/IFR project literature (Chang, 1992b; M.L. Thompson, 1991 and private communication, 1991; K. Wu, private communication, 1991).

Recycle of minor actinides (MA) in an ALMR results in greater decay heat and radioactivity in discharge fuel assemblies. The ALMR project anticipates that the fuel handling equipment, including transport casks, can accommodate the higher levels of decay heat and radioactivity without major design modifications (Thompson et al., 1991).

The ALMR project (Johnson et al., 1990; M.L. Thompson, private communication, 1991) reports preliminary designs for ALMRs fueled entirely with TRUs, with no ^{238}U for breeding or internal conversion. One design is fueled with a mixture of plutonium and separated MAs, with a breeding ratio of 0.22. Another is fueled with MAs, with a breeding ratio of 0.85.[2] Such an MA burner could be useful if separated plutonium were to be recycled to LWRs as mixed-oxide (MOX) fuel—as is being done in Europe and planned in Japan, and if it were found desirable to construct a separate fast-spectrum reactor to transmute the minor TRUs separated in chemical reprocessing of discharge MOX fuel. These designs obviously indicate higher mass depletion rates of TRUs supplied to the liquid-metal reactor (LMR) fuel cycle for start-up and make-up, per unit of thermal power generated, compared with those of the designs of transmutation devices without internal breeding from ^{238}U or ^{232}Th fertile material (e.g., the designs for the accelerator transmutation of waste [ATW] without thorium) considered in the Los Alamos Concepts of accelerator-driven transmutation systems.

However, eliminating internal breeding from fertile material results in shorter fuel residence time in the reactor and lower fuel burn-up in an irradiation cycle. Consequently, the TRU inventories in the external fuel cycle increase, resulting in larger inventories required for start-up before steady-state recycle is achieved.

Effects of the start-up inventories and TRU make-up rates on overall transmutation performance are discussed under *Transmutation Performance*.

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[2] The higher breeding ratio results from neutron absorption in ^{237}Np, producing ^{238}Pu that then absorbs a neutron to form ^{239}Pu.

Transmutation Performance Requirements

Performance requirements for TRU-burning ALMRs have changed markedly in recent years and have not been clearly defined. Varying lines of objectives and arguments have been presented during recent years by DOE and its contractors, ANL and GE. To illustrate the varying lines of argument, in January 1990, DOE (Griffith, 1990) outlined the objectives of the ALMR project with regard to improving waste disposal of spent LWR fuel, stating that the ALMR project expects to develop a new pyrochemical process that will remove 99.9 to 99.999% of the actinides from waste streams, corresponding to process decontamination factors of 10^3 to 10^5.[3] This expectation was said to result in a thousandfold reduction in the quantity of actinide waste going to a geologic repository.

The principal benefit claimed for this reduction in the quantity of TRUs is that the resulting wastes would be expected to be "benign after 200 to 300 years," when the ^{90}Sr and ^{137}Cs fission products will have decayed, reducing the risks from geologic disposal (ANL, 1991; Berglund et al., 1990; Hannum, 1992). ANL (Till, 1990) has stated that the waste from actinide-burning ALMRs "… will not have any actinides in it such as uranium, plutonium or any of the minor actinides such that, after 200 years, the cancer risk to man will be no greater than the original form, i.e., natural uranium." Indeed, if the risks from a given quantity of TRUs in a geologic repository over a given period of time are considered to be serious, one must also consider the risks from significant quantities of TRUs that would remain as yet untransmuted in the ALMR's above-ground fuel cycle—in storage, in reactors, and in reprocessing and fuel refabrication facilities—during the long time period that the transmutation would have to be accomplished.[4]

Apparently, the objective of such a large reduction in TRU inventory in repository waste is based on the ALMR project's desire to reduce the "radiological risk" of the TRUs (Till and Chang, 1989) in a repository to the "cancer risk level" (Berglund et al., 1990) of the original uranium ore mined to fuel the LWRs that generated the spent fuel. The ALMR project apparently assumes that these "risks" are measured by toxicities. The errors in using toxicities and uranium ore comparisons are discussed in Chapter 4.

DOE has clarified some important consequences of the ANL claims. In testimony to Congress in April 1990, DOE stated that the TRU-burning fast reactor "… would reduce the life of the remaining waste to about 200 years, instead 10,000 years, and the waste could then

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[3] Although the transmutation by ALMRs is said to deal with "actinides," the DOE ALMR project actually limits the calculation to transmutation of TRUs. Transmutation of the considerable quantities of uranium recovered from LWR spent fuel is not considered. Because TRU transmutation is spoken of popularly as "actinide burning" within reactor programs in this country and abroad, that terminology is occasionally used herein.

[4] Estimates presented in Chapter 5 show that the time to achieve only a hundredfold reduction in total TRU inventory by the proposed ALMRs would be about 2,800 years for a series of ALMRs of a breeding ratio of 0.65, operating at constant power, assuming sufficient production of TRUs from LWRs to furnish TRU make-up during this period, or about 200 years in a declining-power scenario in which TRU inventory at the end of each reactor life is used to start and fuel a smaller number of subsequent ALMRs. The times for inventory reduction are even longer for other breeding ratios reported by the ALMR project.

be stored above ground until it dies out" (Watkins, 1990). More recently, ALMR proponents have stated that a geologic repository will still be needed for some of the waste streams from the reprocessing cycles, because the process decontamination factor is not sufficiently high to qualify the waste as Class C "non-TRU" waste (i.e., with an alpha activity of less than 100 nanocuries per gram [Till and Chang, 1989]).

Another way of approaching performance requirements is in terms of the DOE schedule (Griffith, 1990) that showed the introduction of 1.4 GWe of new TRU-burning ALMRs in the year 2005, followed by the introduction of an additional 1.4 GWe ALMR per year for the next 40 years. All of the 62,000 Mg of LWR spent fuel accumulated by 2011—the time of opening the first repository—could be reprocessed and used to fuel these ALMRs, together with the yearly discharge of additional spent fuel from LWRs assumed to operate during this 40-year period. By the year 2045, there would be no inventory of LWR spent fuel. However, a geologic repository would still be needed for defense wastes and for the radioactive wastes produced in ALMR operation. The ALMR project does not propose to develop separation processes with decontamination factors sufficiently high that TRU-containing wastes could be disposed of as low-level waste, nor does it propose to transmute fission products present in LWR spent fuel and generated by ALMR operation.[5]

The Department of Energy's 1991 *National Energy Strategy* (DOE, 1991) assumed a similar rate of introduction of TRU-burning ALMRs, beginning in 2012 (Goldner et al., 1991), achieving a level of 19 ALMR power plants by 2030. Each ALMR was assumed to generate 1,395 MWe and to operate at a breeding ratio of 0.6 or 1.01. The *National Energy Strategy* also assumed a growth of total U.S.-installed nuclear capacity from the present installed capacity of about 110 to 195 GWe by 2030, primarily with advanced LWRs, introduced at a rate much greater than that assumed by Griffith (1990). The cases analyzed show that with such ALMR growth, all spent fuel discharged from LWRs from 2012 to 2030 could be processed to fuel the ALMRs, and a portion of the stockpile of LWR spent fuel could also be processed and transmuted during this period.

The new ALMRs would be primarily to treat new spent fuel emerging from LWRs after the first spent-fuel processing plant is built, around 2010 or earlier (Goldner et al., 1991). However, if the ALMRs are available and economically competitive, they could be applied to process and transmute LWR spent fuel that would otherwise be emplaced in the first U.S. geologic repository for high-level waste (HLW). Therefore, if the TRU-burning ALMR program were successful on the schedule expected by DOE and its contractors, there would be no need to load the first U.S. HLW repository with spent fuel (Griffith, 1991). This would call for considerable change in the repository design, in repository safety analysis, in the qualification of waste forms, and in licensing.

Although DOE has not defined performance requirements for the ALMR, the committee finds that the ALMR fuel reprocessing development underway at ANL is aimed at achieving decontamination factors of about 1,000. Therefore, for the purpose of the current study, a

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[5] A 1991 report from ANL (1991) states that fission products are also to be transmuted in the ALMR. However, no information has been provided that shows that the ALMR program is planning to transmute the long-lived fission products that are key radionuclides requiring geologic disposal.

decontamination factor is adopted as a performance goal, and evaluations are made of what overall system performance can be achieved with the various ALMR designs.

Transmutation Performance

While the reactor is transmuting the various isotopes in the fuel assemblies, it is producing higher mass TRUs and fission products in the fuel and blanket assemblies. Calculated annual production and destruction rates of various TRUs in the ALMR are calculated by GE (Thompson et al., 1991) assuming an average fuel burn-up of 150,000 MW·d/Mg, corresponding to a fuel residence time of about 6.5 years.[6]

At the end of the life of an ALMR at constant power, the core and reprocessing inventory would be essentially intact. These could be transferred to subsequent replacement ALMRs to continue power generation and the transmutation. In assessing the performance of the ALMR concept to consume TRUs that would otherwise appear in a geologic repository, it is important to consider the untransmuted inventory in the reactor and in reprocessing. It is also important to compare the total inventory of TRUs in the waste and in the fuel cycle to the amount that would be present for the same power generation from alternative sources, such as from additional LWRs. Calculations presented in the later section, *Comparison of Reactor and Accelerator Approaches* show that several hundred years would be required to reduce the total inventory of TRUs by even a factor of 10 by deploying ALMRs, and thousands of years would be required for a hundredfold reduction, assuming a constant level of nuclear power in the future. The time for a hundredfold reduction could be reduced to a few hundred years if nuclear power were to be terminated as rapidly as possible, consistent with operating a progressively smaller number of ALMRs at constant neutron flux as the reactor and fuel-cycle inventories of the system of several initial ALMRs become transmuted.

Facilities and Support Requirements

To implement the TRU-burning program as described by DOE (Griffith, 1990; Goldner et al., 1991), commercial-scale facilities to reprocess LWR spent fuel would be required. According to the GE ALMR reports, this facility could be an aqueous PUREX/TRUEX process or could be based on ANL's proposal to develop a pyrochemical separation process for LWR spent fuel. GE specifies a reprocessing capacity of 2,700 Mg of LWR spent fuel, more than twice the capacity of any single commercial reprocessing facility. Currently the largest worldwide commercial facility, near completion in the United Kingdom, is the THORP plant, with a throughput of 900 Mg heavy metal. Based on reprocessing/fabrication times estimated by the ALMR project (Taylor et al., 1991), the new commercial-scale LWR fuel reprocessing facility would have to be operational for about 2 years before the commercial ALMRs are

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[6] Fuel burn-up expressed in terms of MW·d/Mg is the average thermal exposure, in megawatt days of thermal energy per megagram (metric ton) of actinides (heavy metal) in the fresh fuel supplied to the reactor.

introduced. Thus, the new large-scale reprocessing facility would have to commence operation by 2003 according to one DOE schedule (Griffith, 1991) or 2010 according to another (Thompson and Gonzaga, 1994).

Argonne National Laboratory's IFR concept of the ALMR specifies that each ALMR power-generating facility would have its own integrated facility for reprocessing discharged ALMR fuel and for fabricating recycle fuel. For a breeding ALMR, the throughput of driver and blanket fuel discharged from an ALMR would be about 21 Mg/yr of uranium and TRUs for each 1,395 GWe ALMR (Johnson et al., 1990). The throughput of ALMR discharge fuel would decrease to about 13 Mg/yr for an ALMR with no uranium blanket. The reprocessing capacity quoted here is that necessary to process the backlog, following DOE's proposed schedule of deployment. Without large-scale reprocessing deployment in the United States, separation and transmutation is an impossible venture.

There must also be facilities for converting the reprocessing waste streams into waste solids suitable for emplacement in a geologic repository or suitable for other waste disposal sites. Necessary processes and facilities, in addition to those described by the ALMR program, include:

- a process to remove actinides and soluble fission products from the inner surfaces of cladding hulls from LWR fuel and a process to produce a suitable waste solid from the cladding hulls;
- a process to remove noble-metal fission products from the uranium stream from pyrochemical reprocessing of LWR spent fuel if that uranium is to be sent to surface storage as now specified by the ALMR program, and a process to produce a suitable waste solid from the separated fission products;
- processes for high-yield (greater than 99.9%) recovery of radioiodine, ^{14}carbon, and technetium if LWR spent fuel is to undergo aqueous reprocessing and processes to produce suitable waste solids from these separated radioelements; and
- processes to separate radioactive cesium and strontium, and to produce suitable waste forms for these separated radioelements, and processes and facilities for storage of these separated radioelements for 200 to 300 years before disposal as radioactive waste (this is a possible additional separation proposed by ANL [Chang, 1992b] and by DOE [Young, 1991] to reduce heat generation in a geologic repository and to increase the amount of waste that could be emplaced in a given repository area).

Qualifying the several different fuel types and compositions that will arise from TRU recycling in the ALMR will be a formidable task.

The ALMR project points out that removing most of the actinides from the HLW would result in a lower rate of heat generation by radioactive decay. This would allow an increase in the total waste loading of a geologic repository of a given area, if that waste loading is limited by the total rate of heat generation. The ALMR program also proposes that the heat-generating fission products ^{90}Sr and ^{137}Cs be separated when spent fuel is reprocessed. By storing the separated strontium and cesium on the surface for about 300 years, the first repository could accommodate more waste than now planned and could defer the time when a second repository is needed (Till and Chang, 1989; Young, 1991). However, Ramspott et al. (1992) point out that the current limits on waste loading in the proposed first repository are not based on heat generation rate. They point out that if the loading were limited by heat generation rate, other techniques could increase waste loading without the complexity of reprocessing and transmutation. If this option is to be pursued, it will be necessary to develop additional separations processes, means of storing the strontium and cesium for hundreds of years, and new waste forms for satisfactory containment of the strontium and cesium for later emplacement in a geologic repository. Containment of the 3-million-year ^{135}Cs is particularly challenging.

Development of the ALMR concept, the new technology for reprocessing LWR spent fuel and for reprocessing and fabricating ALMR fuel, and the additional processes for separating fission products and for preparing special waste forms would require extensive facilities for research, development, testing, and operational demonstration, including prototypes.

Risk and Safety Issues

The National Research Council's (1992) study of nuclear power options for the future summarizes safety features of the ALMR, which is designed only for power generation, and also summarizes risk and safety issues that must be resolved. Two transient safety tests at the EBR-II with metal fuel successfully demonstrated the safety potential of the ALMR concept. The study notes that:

> The properties of the metallic fuel and the large thermal inertia of the sodium pool are key to achieving reactor shutdown passively (i.e., without relying on operation intervention, active components such as control rods, pumps, valves, or the use of balance-of-plant for heat removal) while keeping temperatures low.

However, the report cautions that the presence of a positive sodium void coefficient of reactivity in the ALMR/PRISM design is a continuing concern. It may require revisions to the present design such as a larger diameter core with a lower height.

For ALMRs as TRU burners, GE's approach to eliminating ^{238}U blankets in the ALMR designs of lower breeding ratio requires shortening the core height, with minor adjustment in fuel composition but without changes in fuel-rod diameter or in the number of fuel rods. The diameter of the reactor vessel would remain constant and is limited by the maximum allowable power density or by the linear heat generation rate of the fuel rods so that safety characteristics are not compromised.

With reduced fissile breeding and reduced heavy metal inventory, the burner designs also result in increased reactivity swing over a fuel cycle. This then requires larger control rod worths and hence entails potentially larger positive reactivity insertions and degraded performance in transient overpower events. As the breeding ratio decreases, the positive sodium void coefficient of reactivity decreases, and the Doppler coefficient of reactivity remains negative but becomes smaller in magnitude.

From this approach, GE concludes that a TRU burner with a breeding ratio of 0.60 and a core height of 30 in. is the configuration that would have acceptable safety features and the lowest breeding ratio.

Additional risk and safety issues relevant to the actinide burning, which are identified in the present study, are summarized below. Among the more important issues of reactor and fuel-cycle safety are the following:

1. the effect of reducing the breeding ratio to increase consumption rate of TRUs from an LWR spent fuel, thereby compromising passive safety features;
2. the effect of more favorable neutronic properties of MAs, such as ^{237}Np, on reactivity transients and reactor safety;
3. the effect of the higher specific activity and neutron emission from the decay of higher mass actinides, and from other species resulting from multiple recycle, on radiation exposures and risks from reprocessing, fabrication, and handling of recycle fuel; and
4. the effect on reactor transients and safety if the ALMR is started on enriched ^{235}U instead of starting it with plutonium and other TRUs from LWR spent fuel. The analysis of fuel-cycle economics in Chapter 4 indicates that start-up on ^{235}U would be economically favored if reprocessing costs are comparable to contemporary costs in other countries.[7] The decision as to whether to start ALMRs from LWR fuel transuranics could be further affected if there are important differences in safety features for ALMRs started with transuranics as compared with starting with ^{235}U.

Even though a breeding ratio of 0.60 is said to be the lowest breeding ratio that would result in acceptable safety features, ANL has explored designs with even lower breeding ratio designs, limited to single-batch fueling, with thin fuel pins and loose fuel lattices. The reactivity swing over a cycle can be large, as is especially the case for the burner design fueled with plutonium and MAs, with a breeding ratio of 0.22. For this design, unless the cycle length is substantially shortened, transient overpower events would likely result in unacceptable consequences. The fuel composition is well outside the EBR-II metallic-fuel database. Also, MAs cannot be separated from plutonium in the current pyrochemical processing. Even though the low breeding ratio results in a greater make-up rate of TRUs per unit of thermal power, the large out-of-core inventory for this fuel cycle results in a much lower inventory reduction factor in a given operating time than can be achieved for the PRISM design with a breeding ratio of 0.65.

Fuel consisting primarily of MAs will generate a larger amount of decay heat and may present additional problems in processing and handling. Although the sodium void reactivity worth and Doppler coefficient of reactivity do not appear acceptable for the MA-burner design, there is not sufficient information available for this design to evaluate the reactivity characteristics. Thus, considerable research and design effort appears necessary to develop an

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[7] ^{235}U start-up is estimated (Chang, 1992a) to be economically favored if the unit cost of reprocessing LWR spent fuel is greater than about $350/kg. This assumes uranium ore at $77.8/kg U ($30/lb U_3O_8) and separative work at $80/kg separative work unit, available on the international market.

LMR core that is fueled entirely with TRUs without compromising safety characteristics of current ALMR designs.

The operation of a system using ALMRs can impact the occupational safety and environmental issues associated with the mining and milling of uranium ore, because breeding ALMRs started and fueled with TRUs from LWR spent fuel would require no more fresh uranium ore for the electrical energy generated by these ALMRs. However, if the ALMRs were to operate at a breeding ratio less than unity to increase the amount of TRU burning during a reactor lifetime, then continuing LWRs to supply make-up TRUs, as envisioned in DOE's energy strategy, would require additional fresh uranium ore. Thus, the mixed power system of LWRs and nonbreeding ALMRs would result only in a few-fold decrease in the amount of ore mined per unit of energy produced.

As an alternative to start-up on plutonium, the TRU-burning ALMRs could be started on enriched ^{235}U. If so and if the breeding ratio were unity so that no further make-up ^{235}U were required, the 40-year lifetime requirements of uranium ore for the ALMR would be about the same as for an LWR of the same electrical generation. There would be no significant savings in mining and milling of uranium ore for ^{235}U start-up. However, if the first-generation ALMRs were to operate with a sufficient breeding gain, then the excess plutonium they produce could be used to start next-generation ALMRs, so that no further uranium mining and milling would be required.

A reduction in ore mining and milling from ALMR operation may have only a limited effect on the occupational health and safety of the U.S. nuclear industry within U.S. territory because of two fundamental changes that took place in the industry during the preceding two decades: (1) the adoption of solution mining of uranium in the United States, with an accompanying decrease, compared with conventional mining techniques in the potential for accidents and irradiation; and (2) the large increase in the fraction of uranium ore imported from Australia and Canada, which is projected to range from 72 to 82% of the domestic utility requirements in 1992 through 2002. Indeed, U.S. uranium ore production has dropped by a factor of about 8 since the production peak in 1980, and the drop in employment in the U.S. uranium industry is about a factor of 20 lower than the peak employment in 1979.[8]

TRU-burning ALMRs that are started and refueled with TRUs recovered from LWR spent fuel otherwise destined for a geologic repository would be a means of producing nuclear-generated electric energy without further mining and milling of uranium ore, but this TRU-burning fuel cycle would create a new waste that would be a long-term environmental issue. The proposed flowsheets for reprocessing LWR spent fuel show that uranium recovered from LWR spent fuel would be stored on the earth's surface instead of going to geologic disposal.[9]

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[8] Here the committee must examine the data on the fate of uranium decay daughters (^{230}Th, ^{226}Ra, ^{220}Rn, ^{210}Pb, and ^{231}Pa) that may be brought to the surface by solution mining, as well as the fate of uranium process losses. If an appreciable fraction of the uranium decay daughters and of the uranium itself are left on the earth's surface as waste, the equivalent of mill tailings could be created.

[9] Some ANL flowsheets for pyrochemical processing of LWR spent fuel show that much or all of the long-lived fission product ^{99}Tc would accompany this uranium (McPheeters et al., 1991). Flowsheets developed more recently by the committee and by ANL show that the technetium could be separated and sent to geologic disposal.

About one-sixth of the uranium originally mined to fuel the LWRs that produced that spent fuel would thus become a potential surface contaminant, a contaminant that would present a greater potential risk to future people than if it were buried as a HLW.[10] If this separated uranium, containing about 0.8% ^{235}U, were returned to isotope separation for reenrichment, the amount that would be left for long-term surface storage would be reduced by 20%.

State of the Technology

More than a dozen liquid-metal-cooled fast reactors have been constructed and operated in the United States, in several states in Europe, and in the former Soviet Union. Although some of the reactors have operated extremely well, the overall performance record is mixed. In the United States, two early fast reactors had serious difficulties. The Fermi-I reactor suffered a serious structural failure of certain core components that led to a local flow blockage and fuel melting. The EBR-I test reactor suffered a partial core melt during a dynamics experiment.

The more recent experience in the United States is much more favorable. EBR-II has operated successfully for over 30 years. The larger Fast Flux Test Facility (400 MWt) has had an outstanding record since starting operations in the late 1970s. However, in the United Kingdom, the Prototype Fast Reactor suffered cracking of primary system components as a result of cyclic thermal stresses. France's Rapsodie and the Superphénix liquid metal reactors, as well as the BN-350 reactor in the former Soviet Union, have experienced steam generator leaks. Repair of Rapsodie was considered too expensive to justify keeping the reactor in service after 15 years of operation. The 250 MWe demonstration plant Phénix, which started regular operation in 1974, is shut down pending study and evaluation of transient negative reactivity pulses that were observed in 1989 and 1990 while the reactor was operating at full power. Operation of the Superphénix 1,200 MWe plant has been curtailed for an extended period by a sodium leak, discovered in 1987, in an auxiliary vessel for the storage of discharged fuel (National Research Council, 1992). Future plans for Superphénix appear to be uncertain at present.

The lesson that can be drawn from the experience to date is that sodium-cooled fast reactors can be designed, built, and operated reliably. However, there is a much smaller base of experience, particularly with large LMRs, than with LWRs. There remain sensitive design issues. Proceeding with a program of large-scale TRU burning with an ALMR would require substantial development, testing, and large-scale demonstration under Nuclear Regulatory Commission (NRC) safety review and licensing before one could proceed with confidence. These issues are exacerbated when the proposed new processing technologies are included, particularly if they are included as processes in the IFR concept.

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[10] Even when the uranium in LWR spent fuel is disposed of as HLW in a geologic repository, the long-term risks from the uranium decay daughters is an important environmental issue (Svensk Kärnbärnlehantering AB, 1992).

Almr Fuel

The ALMR is designed to utilize metallic fuel. Its high thermal conductivity affords much smaller temperature swings during reactivity and loss-of-coolant transients. Metal fuel seems better adapted to ANL's proposed pyrochemical processes. Although there is a substantial body of experience with metallic fuel for fast breeders, relatively little of that experience is at high burn-up, and essentially none is with actual recycle fuel containing minor TRUs and the recycled fission products, particularly the lanthanides. The ALMR fuel performance is highly sensitive to the in-reactor microstructure that develops, which is in turn highly sensitive to the phases that precipitate, which are in turn highly sensitive to the chemistry of the fuel. Hence, the in-reactor behavior of a fuel with a significantly large concentration of MAs will need to be extensively characterized. Significant irradiation experience with such fuel will be needed under actual design conditions.

MOX fuel is a possible alternative that has been demonstrated satisfactorily to very high burn-up (greater than 150,000 MW·days/Mg). It seems better adapted to aqueous processing. However, there is essentially no experience with such fuel containing significant amounts of MAs. Also, an oxide-fueled reactor would not have the inherent safety advantage that has been demonstrated for metal fuel. Thus, even with oxide fuel, significant fuel development and irradiation testing would be required. Application of conventional aqueous reprocessing to multiply recycled MOX fuel could increase the necessary cooling time between reactor discharge and reprocessing, further increasing the time to obtain a given reduction in TRU inventory.

Reactor Dynamics And Safety Parameters

Although fast reactors have been operated with mixed plutonium-uranium fuel, there is essentially no operating experience with significant quantities of MAs. Also, there is little experience with recycle fuels containing the lanthanides that would be recycled from pyrochemical reprocessing. In order to provide an adequate basis for licensing, it would be necessary to confirm cross sections, reactivity coefficients, and reactor dynamics over a range of fuel compositions. A representative operating prototype designed and operated under NRC licensing would be required and might also aid in verifying satisfactory operation and design margins at low breeding ratios.

Fuel Fabrication Facilities

The fabrication of fuel containing TRUs will require automated, remotely maintained, and remotely operated fuel production facilities. Such facilities designed for recycle of TRUs recovered from LWR spent fuel and for recycle of TRUs recovered from ALMR spent fuel must be of significant capacity, whether constructed as fabrication facilities colocated with one or more ALMRs, as in the IFR concept, or whether constructed as centrally located facilities with larger throughput. Although there is substantial experience with the fabrication of mixed plutonium-uranium fuels, there are no high-capacity facilities with remote operation and maintenance.[11] The fuel fabrication facilities themselves are likely to require significant development and prototype testing, possibly in support of a research, testing, or demonstration reactor.

If ^{14}C is not removed in reprocessing and made into a suitably insoluble waste form, it can be a troublesome radionuclide in a geologic disposal system. If so, the fuel fabrication processes must maintain a low enough concentration of residual nitrogen in the fabricated fuel to avoid production of significant quantities of ^{14}C.

Reprocessing

Reprocessing would be required to separate the TRUs and fission products from spent LWR fuel as well as from ALMR recycle fuel. As discussed in Chapter 2, there is substantial experience at production scale with the aqueous PUREX process for separating uranium, plutonium, and fission products from spent fuel, including LWR spent fuel. PUREX separation facilities of smaller scale have operated on discharge fuel from LMRs. However, the recoveries are not high enough to meet the process decontamination factor of 1,000 that has evidently been adopted as a requirement by the ALMR program. Also, MAs are usually not separated, except for limited amounts of separations of neptunium. To recover MAs and to achieve high recoveries of all these separated species, the TRUEX process has been studied as an addition to PUREX. TRUEX development has progressed little beyond the laboratory scale. However, the PUREX/TRUEX reprocessing system may be in a more advanced stage of development than the pyrochemical processes proposed for the IFR, including pyrochemical processing of LWR spent fuel.

The ALMR project has proposed pyrochemical reprocessing of discharged metal fuel from an ALMR. Development at ANL is focused toward reprocessing integrated with a reactor complex. Significant experience has been gained on simulated ALMR fuel at a laboratory scale. ANL has also proposed to develop pyrochemical processing technology for high-yield recovery of TRUs from LWR spent fuel, but that technology has not been demonstrated even at laboratory scale (see Chapter 2).

Whatever process or processes are chosen, they will require significant further development and demonstration in a pilot-scale plant of reasonable size, designed and operated under NRC licensing.

It may also be desirable to develop one or more additional reprocessing steps for separation of the long-lived fission products (e.g., ^{99}Tc and ^{129}I) that can significantly affect the calculated risk from geologic disposal. Indeed, the ANL annual report for 1991 (ANL, 1991) states that

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[11] The secure automated fabrication (SAF) line located at Hanford, Washington, was put into operation in the early 1980s to develop MOX fuel for testing in the FFTF as part of the Clinch River liquid-metal reactor project. SAF has a remotely operated and maintained fuel production and nondestructive testing capability with a capacity of about 6 MgHM/year.

> The long-lived fission products are returned to the (IFR) reactor to be consumed. … Long-lived wastes are consumed with reactor fuel, leaving only a small amount of shorter-lived radioisotopes to be disposed of as nuclear waste.

However, the ALMR program has not yet addressed the recycle and possible transmutation of these troublesome long-lived fission products.

Steam Generators

Steam generators have proved to be the most troublesome major component in the pressurized water reactor system. Leaks have been a frequent occurrence; many generator tubes have required plugging. At several reactors, it was necessary to replace the entire set of steam generators. The experience with steam generators in liquid-metal-cooled reactors has been generally better, but the consequences of a leak of high-pressure hot water or steam into sodium are also much more severe. The experience with the EBR-II steam generators has been excellent, with only one leak over their 30-year life. On the other hand, most of the other sodium-cooled reactors around the world have experienced some problems with steam generator leaks.

In the United States, it was envisioned that the Clinch River project would provide experience with large steam generators in a utility operating environment. That program was canceled. Subsequently, DOE maintained a steam generator program for several years, but the program has withered for lack of funding. Because steam generator performance is so important to maintaining reactor availability and capacity factor, pursuit of an ALMR TRU-burning program will require a program for development of large LMR steam generators, with extensive test and demonstration.

Demonstration

To deploy an ALMR system for TRU burning would require reactors, large-scale reprocessing facilities, and fuel fabrication facilities that can operate reliably as a system at high capacity. Because multiple facilities would be required, a TRU-burning program would require predictability of licensing. All of these factors strongly suggest the prudence of prototype or near-prototype demonstration facilities at which the necessary fine tuning of design, the experience base for licensing, and operating experience can be gained. It is not likely, for example, that successful demonstration of pyrochemical processing at EBR-II, as planned by DOE, would provide sufficient basis for reliable design and cost estimating of a large-scale (2,700 Mg/yr) facility for pyrochemical processing of LWR spent fuel, to be funded by risk capital and to be regulated by the NRC. Not only is the process new and unproven even at the laboratory scale, but the capacity is several-fold greater than even the largest commercial reprocessing facility based on the well-developed PUREX process. It is unlikely that the design and safety review for the EBR-II IFR would be sufficient to provide a reliable basis for an industrial design under NRC licensing.

Cost Information

System Costs

Reports of cost estimates made by the ALMR project have been reviewed by the committee. However, the validity of the estimates is uncertain, particularly in the area of capital costs and costs of reprocessing LWR and ALMR spent fuel. For the purpose of this study, the committee relied on the Electric Power Research Institute's (EPRI's) independent estimate of the costs of LWRs and the long-range LMR, as did a recent report on nuclear power by the National Research Council (1992). However, relative costs can be better estimated. It is the cost of the ALMR and its fuel cycle relative to that of the LWR or of an advanced light-water reactor (ALWR) that can reveal the circumstances wherein the ALMR might be competitive as an alternative source of nuclear power and as a means of transmuting TRUs for waste disposal.

The National Research Council (1992) report notes that EPRI has independently evaluated some overnight capital costs and operations and maintenance costs (EPRI, 1989). The estimates are more general than those of the vendors, but they are based on clear definitions. Uncertainties are estimated as -30 to +80%. EPRI's estimates of overnight capital costs are somewhat higher than those of the vendors (National Research Council, 1992). The results for LWRs and ALMRs are shown in Table F-1.

With regard to the relative economic potential of the ALMR, ALMR plants (e.g., PRISM) may be able to compete economically with water reactors if fuel reprocessing turns out to be technically and economically feasible, and if the overnight capital costs of these plants are as low as the vendors indicate (National Research Council, 1992). EPRI costs estimates (Table F-1) suggest that these capital costs will be higher than the vendor estimates.

The economic projections are highly uncertain, first, because past experience suggests higher costs, longer construction times, and lower availabilities than projected and, second, because of different assumptions and levels of maturity among the designs. The EPRI data, which the National Research Council believed to be more reliable than that of the vendors, indicate that the large evolutionary LWRs are likely to be the least costly to build and operate on a cost per kilowatt-electric or kilowatt-hour basis. EPRI puts the midsized LWRs with passive safety features between the two extremes.

However, neither the PRISM design nor the PRISM technology are sufficiently developed to provide a reasonable degree of confidence in cost estimates. Finally, different institutional arrangements may be required for utility involvement in a PRISM plant because of reprocessing, concerns about diversion of sensitive nuclear materials, and lack of utility experience with the technology (National Research Council, 1992).

TABLE F-1 EPRI-Estimated Overnight Capital and Operations and Maintenance Costs (in December 1988 dollars).

		Operations and Maintenance Costs	
Advanced Reactor Type	Overnight Capital Costs (per rated kW)	Fixed[a] ($/kWe-yr)	Incremental[b] (cents/kWh)
Large evolutionary LWRs	$1,300	61.1	0.11
Mid-sized passive LWRs	$1,475	72.7	0.11
Liquid metal reactors	$1,725	75.5	0.15

[a] These operating costs are essentially independent of actual capacity factor, number of hours of operation, or of kilowatts produced. They include labor charges for plant staff.

[b] These variable operating costs and consumables are directly proportional to the amount of kilowatts produced. They include chemicals consumed during plant operation.

SOURCES: EPRI (1989); National Research Council (1992).

The LMR might be commercially competitive if uranium fuel shortages limit the use of LWRs. The LMR's safety features and ability to recycle TRUs are not considered important positive factors for its early market potential. Any strategy requiring fuel reprocessing introduces significant technical, economic, and nonproliferation policy considerations, some of which would complicate licensing.

The TRU-burning ALMR reactor and fuel cycle are expected to be more expensive than an ALMR optimized only for power generation (Chang, 1992; Taylor et al., 1991). Expected contributors to the higher cost for TRU burning are the likely higher cost for high-yield chemical recovery of actinides and fission products; the expense of additional LWR reprocessing for reactors with degraded breeding ratio to increase TRU consumption, and the expense of additional development, testing, and safety issues associated with recycle of MAs and fission products. Therefore, the era of competitiveness of the TRU-burning ALMR with the LWR will be even later than that of economical introduction of ALMRs designed only for power generation.

For the present study, it is important to estimate the time in which the LMR optimized for power generation (and not for TRU burning) could be economically competitive with LWRs. Calculations of the relative economics of ALMR deployment scenarios, involving both ALMRs and LWRs, have been made by Delene, Fuller, and Hudson at Oak Ridge National Laboratory (ORNL) (Delene et al., 1993). At the request of this committee, that analysis has been extended to estimate the future time at which rising ore costs from LWR operation result in a cost of electrical generation from an ALMR that equals that from an ALWR.

The analysis, prepared by C.R. Hudson (private communication, 1993), compared constant-dollar-levelized plant busbar generating cost, including capital, operations and maintenance, fuel, and decommissioning. Relative costs of the power plants were based on the relative overnight capital costs and operating and maintenance costs for large evolutionary LWRs and for LMRs similar to data in Table F-1.[12] All costs except that of uranium ore were assumed to remain constant through time on a real-dollar basis. The ALMR busbar costs were calculated at several different assumed costs of LWR spent fuel reprocessing. Plutonium from LWR spent fuel was assumed to provide the fissile material for the initial core of the ALMR and for the first two reloads. Thereafter, the ALMR was assumed to sustain its own fuel cycle (i.e., unity breeding ratio). Other assumptions used in developing this analysis are

- after-tax cost of money = 9.57%/yr;
- waste fee for ALMR and ALWR = 1 mill/kW·h;
- decommissioning sinking fund charge = 1 mill/kW·h; and
- facilities financed at 70% equity, 17%/yr return on equity, typical of low-risk investment.

ALMR costs are based on recent studies by General Electric (Hutchins et al., 1991) and by ORNL (1992), that adopt ANL's estimates of the cost of pyrochemical processing and fabrication of recycle ALMR fuel.

Figure F-3 shows the calculated levelized busbar cost of electricity from ALWRs and ALMRs as a function of the unit price of natural uranium (dollars per pound of U_3O_8) for various values of the price for reprocessing LWR spent fuel. The busbar costs for the ALWR do not depend on reprocessing cost but increase linearly with the cost of natural uranium. Because the ALMR uses no natural uranium, its busbar cost is insensitive to the ore price but increases with the price of reprocessing LWR spent fuel to obtain plutonium for start-up. LWR spent fuel was assumed to be free. No credit was taken for the value of uranium recovered from LWR spent fuel. The curves do not extend beyond a price of uranium ore of $150/lb, which ORNL estimates to be the cost of uranium from seawater, a resource of large magnitude.

For ore prices as high as $150/lb, the ALMR would be competitive with the ALWR if LWR spent fuel can be reprocessed for about $680/kg. If LWR spent fuel can be reprocessed for $350/kg, the ALMR busbar cost will be greater than that of the ALWR unless the price of natural uranium becomes greater than $105/lb. This is more than fivefold greater than the present price of natural uranium.

A goal of the ALMR project is to develop pyroprocessing LWR spent fuel to cost $350/kg or less, in order to justify starting the ALMR on TRUs from LWR spent fuel instead

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[12] Hudson's calculations are based on a 600 MWe ALWR with a once-through cycle. Precise cost data used in his calculation are to be clarified.

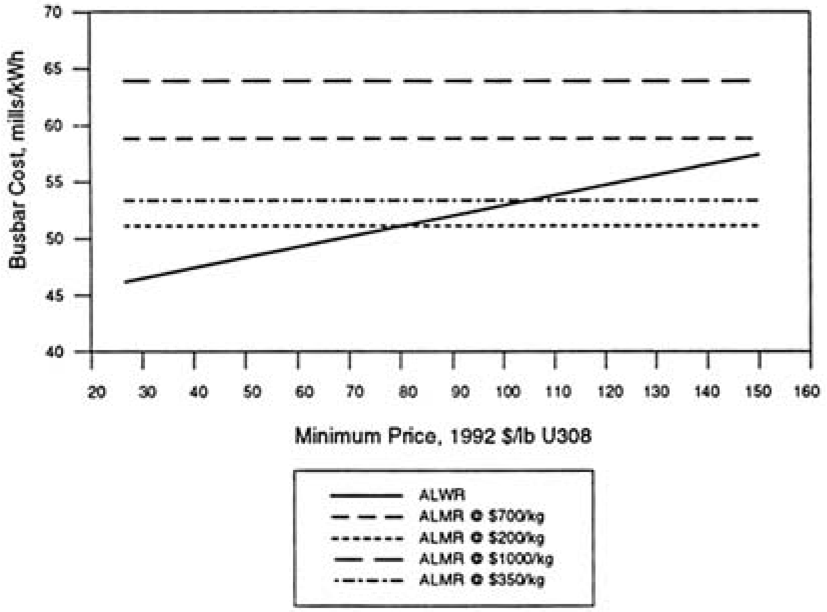

FIGURE F-3 ALMR introduction analysis.

of starting on uranium enriched from natural uranium at an ore cost of $29/lb. From the data in Figure F-3, in an era of low price for uranium ore, as may well occur into the next century, a reprocessing cost even as low as $200/kg would justify starting an ALMR on LWR TURs instead of with enriched uranium, but it would not justify building ALMRs instead of LWRs.

Hudson has estimated the depletion of U.S. uranium resources, assuming only LWRs are added at 1.4 GWe per year during the next century, to be operated in once-through fueling. From data of cost of uranium ore as a function of amount consumed, Hudson then calculated the busbar cost from once-through ALWRs as a function of time, shown in Figure F-4. For a given reprocessing cost, the intersection of this ALWR curve with the corresponding horizontal lines for the ALMR provides the estimated time at which a first-generation breeding ALMR could become as economical as a LWR. If the reprocessing cost is as low as $350/kg (the target value identified for the ALMR project) the time for competitiveness with the LWR is the year 2060. For a reprocessing cost of $1,000/kg or greater, near the estimates for contemporary foreign PUREX plants corrected for U.S. industrial financing, the time of intersection with the ALWR curve would be beyond the year 2070. It may not occur for a far longer time if a large quantity of natural uranium from seawater is available at $150/lb.

These calculations are based on an ALMR plant with a generating capacity of 1,395 MWe, consisting of three power blocks and nine reactors. The fuel cycle is designed to be a break-even cycle requiring fissile material recovered from LWR spent fuel for the initial core and first two reloads. Costs are based on studies by GE (1991) and ORNL (1992). The ALWR plant is a 600 MWe single reactor with a once-through fuel cycle. Capital and operation and maintenance costs are based on vendor input to a U.S. Council for Energy Awareness (1992) study. These cost assumptions are similar to those found in the earlier EPRI *Technical Assessment Guide* (1989). Unit fuel costs are those used in the ORNL ALMR Deployment Economic Analysis (1993).

Busbar costs of electricity include a 1 mill/kWh waste fee and a 1 mill/kWh decommissioning sinking fund charge. All costs are expressed as 1992 constant-dollar levelized values. Constant-dollar levelization is calculated as described by DOE (1988).

A greater capital and operation and maintenance cost for the ALMR will result in even later dates of economical competitiveness, or even lower necessary reprocessing cost. Delene et al. (1993) estimate that if the capital costs of the ALWR and ALMR are equal, the time for equal generation costs would be about the year 2060, assuming a reprocessing cost of $1,000/kg.

The TRU-burning ALMR reactor and fuel cycle are expected to be more expensive than for an ALMR optimized only for power generation (Chang, 1992b; Taylor et al., 1991). Expected contributors to the higher cost for TRU burning are the likely higher cost for high-yield chemical recovery of actinides and fission products, the expense of additional LWR reprocessing for reactors with degraded breeding ratio to increase TRU consumption, and the expense of additional development, testing, and safety issues associated with recycle of MAs and fission products. Therefore, the era of competitiveness of the TRU-burning ALMR with the LWR will be even later than that of economical introduction of ALMRs designed only for power generation, unless the TRU-burning operation is subsidized by possible improvements in performance of a geologic repository. In the absence of considerable subsidy for possible

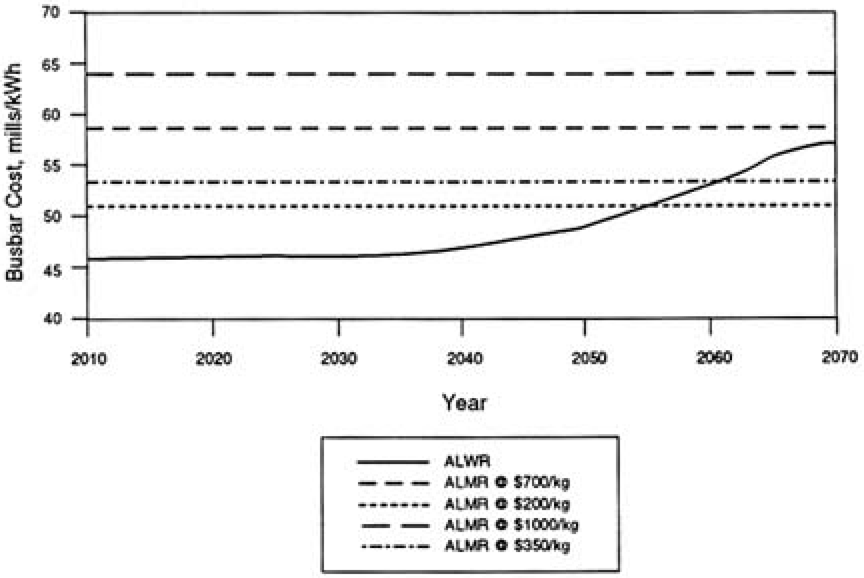

FIGURE F-4 ALMR introduction analysis (Using ORNL deployment data).

waste-disposal benefits, the TRU-burning ALMR is not likely to be an economic choice for power generation until well into the second half of the next century or later.

The Light-Water Reactor

Plutonium Recycle in LWRs

Light-water reactors are by far the most mature of any of the proposed burners of nuclear waste. Over 100 LWRs are operating in the United States and about 400 total worldwide. LWRs have over 4,000 reactor-years of operation and supply about 70% of the electricity consumed in France and some parts of the United States. Fresh fuel for an LWR normally uses low-enriched uranium (LEU) as the fissile component. However, operation is also possible with MOX fuel, which combines plutonium, recovered from reprocessing, with natural or LEU oxide as a PuO_2-UO_2 mixture. This section discusses an extension of the experience with such plutonium-uranium MOX fuel in LWRs to the more complex requirements for refabricating and burning MOX fuel that contains radionuclides recovered from reprocessed LWR spent fuel (i.e., plutonium, the MAs, and selected fission products ^{99}Tc and radioiodine).

The U.S. Plutonium Utilization Program began in 1956 and stimulated related work in several European nations and Japan. This development effort was motivated by the potential for fuel-cycle economies through the recycle of plutonium recovered from LWR spent fuel in anticipation of the rising costs of LEU in the future. In the mid-1960s, the United States conducted a commercial recycle demonstration at San Onofre including a full core load of MOX fuel. Several U.S. reactors conducted tests with partial core loadings of MOX fuel.[13]

Other nations have also successfully operated commercial LWRs with MOX fuel on a limited scale for several decades (Bairiot, 1984; Bairiot and Vandenberg, 1989). Beginning in 1963, Belgium used MOX fuel in their BR3 pressurized water reactor (PWR), achieving a proportion of 70% MOX fuel in 1986. Germany started the commercial use of MOX fuel in 1981, following extensive test and demonstration from 1968 to 1977, and licensed a large MOX fuel fabrication plant at Hanau, although construction was cancelled in 1992 after design was completed. Switzerland began using MOX fuel in the Beznau reactor in 1984, and Electricity de France began loading MOX on a commercial basis in 16 plants in 1985. Japan launched a MOX demonstration program in 1986 and currently is considering the use of MOX fuel in 12 PWRs and several boiling water reactors (BWRs) in the 1,000 MWe range. Indeed, France, Germany, and Japan have the capability for extensive MOX fuel programs in the future, depending whether planned or projected MOX fuel fabrication facilities are actually constructed and put into operation.

Work on reprocessing and plutonium recycle came to an end in the United States in the mid-1970s, as a result of both economic and public policy considerations that are discussed in

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[13] For a history of the U.S. Plutonium Utilization Program, details on the results obtained, and the issues raised by plutonium recycling, see GESMO (1975); see also Leggett and Omberg (1987).

Chapter 1 and 4. The increased availability of enrichment capability and low-cost uranium ore eliminated the economic incentive for plutonium recycle in LWRs and postponed breeder commercialization. Also, the concerns about the possible stimulus that widespread commercial plutonium recycle might give to the proliferation of nuclear weapons played a significant role in the decision of the United States and several other nations to forego recycle. However, several nations—notably, France, the United Kingdom, Russia, and Japan—continue to pursue reprocessing for limited recycle of plutonium into LWRs and for support of their ongoing breeder development programs.

To meet the objective of transmuting the TRU and long-lived fission product inventories as a part of reducing waste disposal risks, the LWR with aqueous reprocessing is the most mature option technologically, requiring the least development time and effort. Present-day LWR designs are readily adaptable for the task. In contrast, fuel-cycle economics with reprocessing is far less certain. For fresh fuel containing radionuclides, the fuel development and the fuel and system performance verification would require time to obtain a database sufficient for licensing, as discussed below. Time also would be needed to construct and license the necessary fuel reprocessing and refabrication facilities in the United States. Thus, advanced LWR designs, which are currently under development and regulatory review, could be alternative candidates to present-day LWR designs to adapt for waste transmutation. In particular, the designs might be useful as the dedicated TRU burners discussed in the next subsection.

Reference Designs

Self-Generated Recycle With Transuranics

In the mid-1960s to the mid-1970s, the U.S. nuclear industry adapted the PWR and BWR designs for "self-generated uranium-plutonium recycle" in which the fissile material recovered from reprocessed LWR spent fuel would be recycled back to the same reactors. Figure F-5 shows a representative material flowsheet for a commercial 1,000 MWe PWR based on a design of mid-1970s vintage by Combustion Engineering[14] (Pigford and Yang, 1977; Hebel et al., 1978). Other LWR designs would have comparable flowsheets for self-generated recycle.

Self-generated recycle schemes typically use heterogeneous cores that contain both rods with normal LEU (about 3.3% ^{235}U) fuel and rods with MOX fuel, so that the reactor characteristics are very similar to those for normal fueling. Two arrangements have been used to accommodate the two types of fuel materials and maintain a suitably flat power distribution in the core. Fuel rods are packaged in "fuel assemblies" that contain arrays of 14 × 14 to 18 × 18 fuel rods for a typical large PWR. In the first arrangement, each assembly may have both

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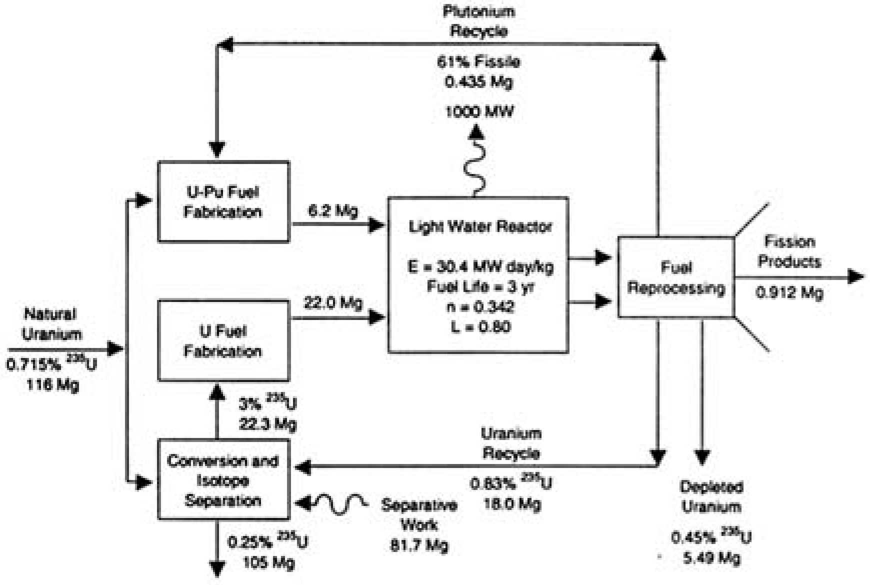
[14] Now Asea Brown Boveri Combustion Engineering Nuclear Power. The data for self-generated recycle is published in Shapiro et al. (1977). The material balance of Figure F-5 would need to be altered for the material flows required in a system to accommodate the already existing LWR spent fuel.



SOURCE: Pigford and Yang (1977).

FIGURE F-5 Material flowsheet for pressurized water reactor with self-generated uranium-plutonium recycle.

normal rods and MOX rods. In the second arrangement, each assembly contains rods only of one type or the other. Both arrangements have advantages and disadvantages.

The use of plutonium introduces new issues of reactivity control and power distribution in the reactor because of the higher neutron-absorption cross section for ^{239}Pu compared with ^{235}U. Thus, the distribution of neutron flux and the concentration of plutonium in MOX rods are critical design issues that establish how the two types of fuel rods are distributed in the fuel assemblies. In the first arrangement, a MOX fuel rod near normal fuel rods, exposed to nearly the same neutron flux, may generate more thermal power. Thus, the proportion of the two types of rods may be varied depending on where an assembly is to be placed in the reactor (at the interior, periphery, or corners of the core).

The first arrangement gives a good match to the local power generation and simplifies the reactor operator's task but complicates fuel assembly production and testing. The second arrangement, in which an assembly contains rods of only one type, simplifies the task of the fuel producer. However, in a MOX assembly there may be considerable variation in flux from the assembly surface to its interior, which requires attention to the concentration of plutonium in the various rods. Thus, the second arrangement complicates the task of the reactor designer and operator to ensure that all fuel rods operate at the desired power within safety limits.

Moreover, the higher absorption cross section in ^{239}Pu reduces the effectiveness of the usual reactor control absorbers. For self-generated recycle of plutonium containing mainly ^{239}Pu, designers concluded that the existing control absorbers could be used if no more than about one-third of the core were fueled with MOX fuel. A larger loading would require modification of the pressure vessel head in a PWR (or the bottom-mounted control structure in a BWR) to accommodate more control rods.

Adapting the self-generated recycle approach to the transmutation of the MAs and selected fission products could proceed by mixing these constituents with the plutonium in the MOX fuel rods. The addition of MAs and ^{238}Pu, ^{240}Pu, and ^{242}Pu complicates somewhat the issues of reactivity control and core power distribution, due to the changing composition and reactivity of the waste material to be burned as the transmutation proceeds over many fuel cycles. In particular, the isotopes ^{241}Am and ^{240}Pu have large cross sections for thermal neutrons and significantly affect the reactivity. However, recent work (Lee and Du, 1994) confirms earlier studies that show that self-generated recycle with MAs and TRUs is could be feasible with one-third core loading. Moreover, the changing fuel composition and the presence of very high actinide radioactivity in discharged and recycled fuel complicates fuel reprocessing and fabrication, quality assurance, and fuel-performance testing and verification that are required for licensing. These reactivity control and fuel issues are discussed more fully under *State of the Technology* below.

Dedicated LWR Transuranic Burners

The U.S. nuclear industry also envisioned an alternative approach to self-generated recycle with one-third core loading. Some of the PWRs were designed with the flexibility of adding enough control absorbers so that these reactors could be fueled entirely with MOX fuel containing natural uranium rather than LEU. These LWRs would operate as dedicated "plutonium-burner" reactors while the other LWRs would be fueled entirely with normal LEU. This would avoid the problems arising from two types of fuel rods with different neutronic characteristics. To flatten the power distribution in the core, the concentration of plutonium in the rods may be varied depending on where an assembly is to be placed in the reactor. Thus, the division into plutonium burners and normal LWRs would simplify the issues of control-absorber design, reactivity control, and local heat generation rate in the MOX fuel rods. Such plutonium burners could serve as devices for transmuting the MAs and fission products.

Any of the current or advanced LWR designs could be adapted for such a purpose. Figure F-6 shows a representative material flowsheet of a plutonium-burner PWR based on the same commercial 1,000 MWe PWR design by Combustion Engineering as for self-generated recycle in Figure F-5 [15] (Pigford and Yang, 1977; Hebel et al., 1978). An LWR is an efficient alternative for dedicated plutonium burning because it can take advantage of the high fissile specific power of a thermal reactor that results in a high fractional burn-up in the target fuel.

However, the changing composition and the radioactivity from the MAs and the various plutonium isotopes in the target fuel material raise similar issues—concerning fuel refabrication, quality assurance, and fuel performance testing and verification—for an LWR TRU burner as is discussed above for LWR self-generated recycle. The recent study of Lee and Du (1994) indicates that the neutron absorption by the ^{241}Am and ^{240}Pu isotopes may build up to levels in dedicated burner operation that requires addition of uranium of moderate enrichment (a few percent) to maintain reactivity. This would also enhance the capabilities for burning the long-lived fission products.

Advanced LWR Designs

In recent years, the EPRI has conducted a program for U.S. utilities to reconceptualize LWR designs based on the extensive LWR experience in the United States and abroad. The overall objective is to achieve greater engineering and physics margins and reduced complexity of construction and operation, compared with previous LWRs. Two classes of designs are in the process of being certified for standardized licensing by the NRC. The goals are at least a factor of 10 reduction in the potential for any core-damaging accident and an even greater reduction in the potential for public harm. Additional goals address better economics.

Specific designs for "large evolutionary LWRs," all rated about 1,300 MWe, have been proposed by Westinghouse for the advanced PWR-1300, by GE for the advanced BWR, and by Asea Brown Boveri Combustion Engineering Nuclear Power for the System 80+ PWR. These systems generally conform to requirements set forth in the Utility Requirements Document (EPRI, 1990), summarized in Table F-2 and serve as benchmarks

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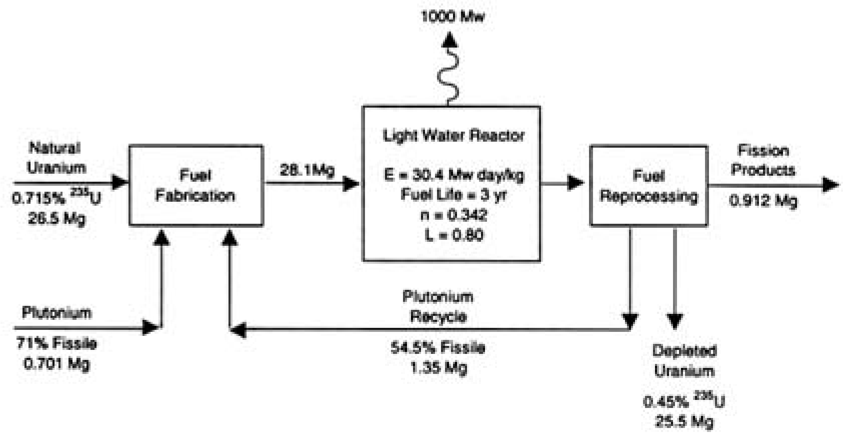
[15] The data for the plutonium burner is published in Shapiro et al. (1977). The flowsheet and material balance of Figure F-6 would need to be altered to include the MA and fission product feeds.



Source: Pigford and Yang (1977)

FIGURE F-6 Material flowsheet for uranium-plutonium fueled pressurized water reactor.

TABLE F-2 Key Utility Design Requirements for Advanced Light-Water Reactors [a]

Plant size	Reference size 1,200–1,300 MWe for evolutionary designs; reference size 600 MWe for passive safety designs		
Design life	60 years		
Design philosophy	Simple, rugged, no prototype required		
Accident resistance	≥ 15% fuel thermal margin, increased time for response to upsets		
Core damage frequency	< 10^{-5}/yr by probabilistic risk analysis		
Loss of coolant accident	No fuel damage for 6° pipe break		
Severe accident mitigation	<25 rem at site boundary for accidents with > 10^{-4}/yr cumulative frequency		
Emergency planning zone	For passive plant provide technical basis for simplification of offsite emergency plan		
Design availability	87%		
Refueling interval	24 months capability		
Maneuvering	Daily load follow		
Worker radiation exposure	< 100 person rem/yr		
Construction time	1,300 MWe: ≦ 54 months (first concrete to commercial operation); 600 MWe: ≦ 42 months		
Design status	90% complete at construction initiation		
Economic goals	10% cost advantage over alternative (nonnuclear) after 10 years and 20% advantage after 30 years		
Resulting cost goals (1989 dollars)		1,200 MWe[b]	600 MWe[b]
	Overnight capital 30-year levelized total generation	1,300 $/kWe 6.3 cents/ kW·h	1,475 $/kWe 7.2 cents/ kW·h

SOURCE: Electric Power Research Institute (1990).

[a] These requirements apply to both the large evolutionary LWRs and to the mid-sized LWRs with passive safety features.

[b] 1,200 MWe commercial operation in 1998; 600 MWe in 2000.

for the near-and mid-term future. The design improvements cover every facet of the systems—reactor core design, fluid systems, steam generators, control and instrumentation, and containment.

In addition to the evolutionary improvements in design, GE and Westinghouse have proposed LWRs of 600 MWe that would employ primarily passive means—gravity, natural circulation, and stored energy—for the main safety functions. Known as "mid-size LWRs with passive safety features," such reactors could be attractive because of the simplification in system components (tanks, valves, pipes, cabling, ducting, etc.) with accompanying reduction in cost and improvement in operability, maintainability, and reliability (National Research Council, 1992:102–110). Also, such reactors may reduce the reliance on active components and human operators, relative to present-day designs, to cope with the wide variety of abnormal conditions. The passive safety features may even eliminate the need for special safety systems, such as the emergency core cooling system in present-day designs.

The Westinghouse AP-600 design and GE's simplified boiling water reactor design presumably require more development before commercial deployment than would the large evolutionary LWR designs. Nonetheless, the passive safety designs potentially could be available for start of construction by the end of this decade, and the evolutionary designs by the mid-1990s, depending on the pace of licensing and utility interest. Indeed, an AP-600 has been proposed recently as a weapons plutonium burner (Buckner and Parks, 1992). However, the critical issues raised above for the current designs—concerning reactivity control and local heat generation rate in the MOX fuel rods, as well as radioactivity in the refabricated fuel—would impact transmutation with either class of advanced LWR designs.

Transmutation Performance Requirements

The MAs could be burned in a thermal spectrum. Also, LWRs could transmute the technetium and iodine fission products. The estimated fractional burn-up rates are 11 and 3%/yr, respectively (Croff et al., 1980, 1990). In addition, the LWRs could furnish make-up plutonium if a transmutation system has other reactors that act as waste burners and are net plutonium consumers.

However, the higher capture-to-fission ratio for a thermal spectrum results in a greater build-up of high mass MAs. Successive neutron captures would produce several curium isotopes that decay by spontaneous fission and produce fast neutrons. Some will decay by alpha emission and produce enough heat to produce a cooling problem (see Chapter 4). Much of the curium alpha and neutron activity would come from ^{242}Cm and ^{244}Cm with contributions from ^{246}Cm and ^{248}Cm as these isotopes build up in later fuel cycles of the transmutation period. Further capture of neutrons together with beta decay would produce some ^{252}Cf, which is an intense emitter of neutrons from spontaneous fission. In addition, unburned ^{238}Pu would be a significant neutron source and heat producer. Thus, the build-up of high mass TRUs over many fuel cycles poses an issue for fuel reprocessing, target fuel refabrication, quality assurance, and fuel performance verification (see *State of the Technology* below).

In the residual high-level waste (HLW) after LWR transmutation, the curium isotopes and ^{238}Pu may build up to very high activities, sufficient to dominate the short-term risks from a geologic repository. The ^{234}U and ^{226}Ra decay daughters of ^{238}Pu would add to the long-term risk. Transmuting ^{237}Np would not necessarily reduce its long-term risk appreciably if neptunium is solubility limited. Transmutation of ^{129}I and ^{99}Tc, or deploying better repository waste forms for these radionuclides after chemical separation, could significantly decrease the calculated individual dose and risk from a repository. However, using transmutation as a means to add more waste from future nuclear power systems to the proposed Yucca Mountain repository, as DOE has proposed, could increase the individual doses and risks from that repository, even for radionuclides that have been reduced in inventory by transmutation. Predicting the resulting effect on repository performance would depend on detailed calculations that have not yet been performed.

The comparative transmutation performance is discussed in Chapter 4, where inventory reduction factors are calculated for the Combustion Engineering PWR design as a plutonium burner, assuming a process decontamination factor of 1,000. By comparison with an ALMR of similar reactor breeding ratio (i.e., 0.65), a higher asymptotic inventory reduction factor seems possible with the LWR—about 4,800:1 versus about 3,000:1. Similarly, the time required to attain a given inventory reduction would be less than for an ALMR.

The superior performance of the LWR plutonium burner results from the higher fissile specific power of an LWR reactor, which should prevail even when MAs are included. The comparative performance is similar, but time scales are shorter by a factor of about 5 for a declining power scenario in which nuclear power is phased out as rapidly as possible consistent with transmuting the maximum amounts of TRUs and key fission products in the HLW.

Facilities and Support Requirements

Reprocessing Requirements

Transmutation with LWRs requires facilities of high reliability and capacity factor to reprocess the LWR spent fuel. Although the reprocessing of commercial reactor fuel was discontinued in the United States in the mid-1970s, a facility for reprocessing commercial reactor fuel had operated at West Valley, New York, for a short period, and two large commercial reprocessing facilities had been constructed at Barnwell, South Carolina and Morris, Illinois, but not operated. These plants were based on aqueous PUREX technology to separate uranium and plutonium from HLW and were designed for contact maintenance after the PUREX initial separations stage. Also, they were not constructed with international safeguards in mind. Thus, their design experience is relevant, but their costs would not fully reflect present-day requirements.

With aqueous-based technology, the facility could use a TRUEX process stage after the basic PUREX process for better separation of the MAs and reduction of the TRUs in the various waste streams to as low a level as possible. Additional processing might be employed after the TRUEX stage to separate the technetium and possibly other fission products. Also, radioiodine would be recovered from the off-gases. TRUEX has been demonstrated on actual reactor waste at laboratory scale, but a process scale-up and pilot-scale test/demonstration would be necessary with waste containing MAs. It may become possible to use pyroprocessing technology for LWR waste, but its feasibility has yet to be demonstrated for oxide fuels, even at laboratory scale (see Chapter 2).

Based on trends in the United States and overseas for increased protection of operational personnel as well as the general public, one could assume the requirement for a fully automated, remotely maintained type of reprocessing facility (i.e., with heavy shielding and equipped with sufficient automation and redundancy to avoid the need for human access even for maintenance). Such a facility presumably would include automated materials accountability and advanced surveillance capability for safeguards.

Around 1980, a detailed study was conducted by Bechtel and Oak Ridge National Laboratory (ORNL) for a plant with all-remote operation and maintenance for reprocessing oxide breeder reactor fuel, including provision for safeguards (Jones, 1981). In addition, the experience of foreign commercial reprocessors using aqueous-based separations is highly relevant. A large reprocessing facility (UP2) at La Hague, France, with a dual defense/commercial mission has operated for over a decade, and a commercial facility (UP3) with a nominal operating capacity of 800 MgHM/yr has recently been constructed. The United Kingdom operated a small reprocessing facility at Dounreay, Scotland, and is constructing a commercial plant (THORP) of 800-MgHM/yr nominal operating capacity at Sellafield. Japan has operated a small reprocessing facility at Tokaimura and is planning for a facility at Rokkashomura of 800-MgHM/yr nominal operating capacity (see Chapter 4 and Chapter 6, *Cost of Fuel Reprocessing*). However, the European plants are not designed for totally remote maintenance. The design of the planned Japanese facility at Rokkashomura may be closer to the anticipated requirements for a U.S. facility.

Thus, a considerable base of experience exists on which a commercial-scale reprocessing facility could be designed and operated in the United States to meet present-day requirements. A plant the size of UP3 or the planned Japanese reprocessing facility (nominally 800 MgHM/yr operating capacity) could support many LWR TRU burners. In addition to technical issues such as TRUEX development, however, there are difficult issues of proliferation policy, public acceptability, licensing, high capital costs, and overall fuel-cycle economics. The development and system costs are estimated below. For other issues, see Chapter 6.

Fuel Fabrication Requirements

In addition to reprocessing capability, one or more facilities would be needed for MOX fuel fabrication, which might be collocated with reprocessing. (The technology is discussed below under *State of the Technology*.) Because of the much higher level of radioactivity in the fuel material from transmutation, relative to LEU, the fuel refabrication would require a fully automated MOX facility constructed for all-remote maintenance. Automated materials accountability and advanced surveillance capability for safeguards would presumably also be a requirement (Pillay et al., 1987). The closest to such a facility in the United States is the secure automated fabrication (SAF) line in the fuel cycle plant at Hanford, Washington.[16] The SAF line has a nominal capacity of 6 MgHM/yr (on a single shift) with provision for extensive fuel materials evaluation capability. Nondestructive test capability includes, for example, the use of neutron irradiation from a ^{252}Cf source to assure uniform distribution of fissile material in fuel rods. The facility also provides for a fully automated materials accountability system as the keystone of a safeguards system.

A recent design for a much larger MOX fabrication plant is that of the Siemens Mixed Oxide Fuel Fabrication Facility at Hanau, Germany. The specification is for a facility of nominally 120 MgHM/yr at a cost reported to be about $500 million (*Nuclear Fuel*, 1992). The Siemens plant "… incorporates a high degree of automation and remote control, remote handling of plutonium for MOX fuel, and increased radiation protection of manufacturing personnel" (Schmiedel, 1991). The plant would have capability for sophisticated materials characterization and testing to produce fuel assemblies optimized for use of plutonium. The last partial construction license, which includes the operating license, was issued in March 1991, with the anticipation that the new facility would eventually replace the current Siemens MOX fabrication plant. After strong opposition by the public, however, the state government reconsidered the project that was cancelled in 1992.

A recent proposal to the Department of Energy, one of several for fuel-cycle systems to support the possible burning of plutonium from dismantled nuclear weapons, specifies a 150,000-ft^2 fully automated MOX facility with a nominal capacity of 60 to 70 MTHM/yr, which could support up to nine AP-600 reactors operating on self-generated recycle (Buckner and Parks, 1992). (See discussion under *System Costs* below.) The facility would provide for powder conversion, pellet production, fuel assembly fabrication, inspection, and shipping. The preliminary plan has parallel lines for redundancy and could be expanded for additional capacity. A detailed design of the plant is not yet available, but its automation philosophy may offer a useful point of reference for the type of MOX fabrication plant needed to support LWR transmuters.

Residual Waste Management and Environmental Impacts

Waste packaging and disposal will be required for any HLW arising from fuel reprocessing and fabrication during transmutation and the residual HLW remaining at the end. It could be packaged (vitrified) with the shorter-lived fission products (mainly ^{90}Sr and ^{137}Cs) and disposed in the geologic repository required for those fission products and miscellaneous TRU-contaminated trash. The estimates of low-level waste (LLW) and other waste streams are not complete enough as yet to set requirements for the packaging and disposal of those constituents.

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[16] The SAF line was built in the early 1980s at a cost of about $100 million (including research and development) to fabricate MOX breeder fuel for test and performance verification in the nearby FFTF. After the cancellation of the Clinch River program, SAF has not undergone "hot" operation.

State Of The Technology

LWRs are at a rather advanced state of development, especially in relation to the other conceptual transmutation approaches considered in this report. Nonetheless, for the LWRs and the associated fuel cycle for transmutation, the development issues include the following:

1. *Verification of control absorber effectiveness and reactivity control*, to offset the greater neutron absorption due to the higher concentrations of TRUs compared with normal LWR fuel. This includes matching the power from fuel rods of different and changing compositions and, for a dedicated plutonium burner, possible addition of uranium of moderate enrichment to maintain reactivity. The extensive work to date here and abroad on MOX fuel furnishes a solid basis for the additional work required with transmutation target fuel.
2. *Safety studies and licensing acceptability of LWRs with TRU fuels* , which would include the review of a generic environmental impact statement on MOX fuels. This would reopen and conclude the process begun with the Generic Environmental Statement on Mixed Oxide (GESMO) fuel hearings terminated in the mid-1970s, a requirement also faced by the other waste transmutation options. Safety qualifications would be required on the specific fuel systems, as well as licensing of the reprocessing and fuel fabrication facilities. Shipping and storage of MOX fuels also must be addressed.
3. *Development and pilot-scale test and demonstration of a complete separations process for the spent fuel, with minimal generation of HLW*. For an aqueous-based process, this would include scale-up and pilot-scale demonstration/test of the TRUEX process. For a pyrometallurgical process for LWR spent fuels, laboratory-scale development must be completed prior to further scale-up and demonstration/test.
4. *Development, quality assurance, and performance verification for fuels* that contain changing amounts of TRUs and fission products. This includes the capability for long burn-up, with a database and fuel specifications sufficient for NRC licensing. The effort entails the refinement and demonstration of nondestructive testing techniques that can remain effective in the presence of the radioactivity in the refabricated fuel.
5. *Verification of a highly reliable, remotely maintained and operated manufacturing process for fuels with TRUs and fission products*. The process and the fuel reprocessing and refabrication facilities would have to meet the current U.S. standards for occupational (and public) health and safety, which are tighter than those that prevailed for U.S. facilities built in the 1960s and early 1970s. The licensing process may be time consuming because of the lack of recent regulatory history and regulatory guides for reprocessing and refabrication in the United States. In addition, assay techniques must be developed, demonstrated, and accepted by regulatory bodies for determining the elemental and isotopic inventories at each stage of the fabrication process. The techniques would have to be integrated into an accountability system that meets safeguards and NRC requirements and is consistent with reliable plant operation at a high capacity factor.

6. *Development and testing of a packaging process for the residual HLW* from an LWR transmuter and its fuel cycle, suitable for licensing by the NRC for geologic disposal. The other waste transmutation options also face this requirement.

Reactor Control Absorber And Reactivity Control Issues

Several issues affecting reactor safety and licensability arise from the larger neutron-absorption cross section and the lower number of neutrons produced per absorbed neutron for ^{239}Pu compared with ^{235}U, as outlined above in *Reference ALMR Designs*. Although the fissile concentration of the plutonium is about the same as in normal uranium fuel, the larger cross section for plutonium results in less effectiveness of the control absorbers. Additional effects would arise from neutron absorption and fissioning in MAs that would build up during the long transmutation period. However, plutonium would still be the dominant TRU throughout the many fuel cycles, assuming that make-up fuel material is available as under "steady nuclear power" scenarios.

For self-generated recycle, the designs compensate by adding control rods or by limiting the amount of plutonium and MAs. Because of the higher thermal fission cross section for ^{239}Pu, there is a tendency for power peaking at or near the MOX fuel rods. The spatial distribution of neutron flux and of plutonium concentration will vary considerably during the time (several years) that an assembly is irradiated in the reactor. The composition of nearby uranium fuel also varies but in a different way than does the MOX fuel. Thus, for self-generated recycle (with or without actinides), it is a complicated design problem to ensure that all fuel rods operate at the desired power within safety limits during the irradiation period.

For the dedicated plutonium burner approach, some of these design issues are considerably simplified, but the price is an increased number of control rods and the need for the addition of some uranium of moderated enrichment. However, the annual MOX fuel replacement for such a reactor could be significantly greater than would be possible with existing reactors under a self-generated recycle approach. Consequently, the plutonium burner approach may be the more flexible of the two when the complication of MAs is imposed on the reactor. Nevertheless, the self-generated recycle approach with existing reactors would be feasible. The computational codes and isotopic cross sections exist to evaluate the reactivity control issues and optimize the performance for either approach.

In addition to the usual change in the concentrations of isotopes during irradiation, noted above, there will be a gradual secular change in the concentrations of TRUs as the burning proceeds. This will require careful balancing of fissile components and control absorbers to maintain suitable operation over the transmutation period. One would expect the insights and techniques gained in the study and demonstration of plutonium recycle for several decades to carry over to the situation with MAs (and technetium and radioiodine) present in the fuel. However, the program must plan for an extended period of in-reactor test and demonstration to build the database that would be required to operate an LWR TRU burner of either the self-generated recycle or dedicated burner type over the many fuel cycles.

Fuel Fabrication And Performance Verification Issues

The target fuel for transmutation, containing TRUs and selected fission products, would draw on the experience with conventional MOX fuel in LWRs and fast reactors (U.S. Nuclear Regulatory Commission, 1975; Leggett and Omberg, 1987; Bairiot and Vandenberg, 1989; Buckner and Parks, 1992). Both mechanical blending of plutonium and uranium oxides and coprecipitation have successfully produced MOX fuel material. The U.S. effort involved extensive small-batch fabrication and testing, with commercial fabrication of MOX fuel elements up to the mid-1970s by two manufacturers—Kerr McGee at a plant in Oklahoma and the Babcock and Wilcox Nuclear Materials Division at a plant in Pennsylvania.[17]

More recently, DOE funded a major program of MOX fuel development and performance verification to support the development of the Clinch River reactor, emphasizing fuel reliability at high burn-up as a key to an economical fuel system (Leggett and Omberg, 1987). The overall experience with ceramic, metal, cermet, and MOX fuels illustrates the need for stringent quality control and underscores the exacting nature and significant cost of the comprehensive fuels development, testing, and performance verification that is required for licensing, not only for LWRs but for any transmutation system.

One of the fundamental advantages of the thermal spectrum associated with LWRs is the ability of destroy several of the water-soluble, long-lived fission products that tend to dominate calculated long-term repository risk. However, there has been little development of fuel materials and target forms in which the fission products may be irradiated. Significant development, demonstration, and testing would be required for a licensable wasteform.

Also, as noted above, the fuel would have secular changes in isotopic concentration over the long transmutation period from the build-up of higher actinides.[18] Moreover, issues with fuel inhomogeneities and degradation, which can occur with any nuclear fuel system, need to be worked out for MOX fuels containing the MAs and selected fission products. Degradation issues can arise from impurities (e.g., flourine, chlorine, water, carbonaceous material), variations in oxide–metal ratios, and cladding characteristics (e.g., grain size, orientation, lining). Fuel fabrication processes also can exhibit problems over the range of TRU composition. For example, the MOX coprecipitation process exhibits the potential for segregation of the plutonium into plutonium-rich areas. The mechanical mix-blend process exhibits a tendency of PuO_2 to resist fine-scale blending with the UO_2.

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[17] These plants used different MOX fabrication processes. The Kerr McGee plant used coprecipitation to form the mixed oxides, whereas the Babcock and Wilcox plant prepared the oxides of uranium and plutonium separately, mechanically blending the two oxides prior to calcining and pressing the pellets. The flowsheets for the plants are similar; the main differences are in the head-end treatment (U.S. Nuclear Regulatory Commission, 1975; Hebel at al., 1978).

[18] Such secular changes in concentration would occur even for a "steady nuclear power" scenario; the range of composition could be wider still under a "declining nuclear power" scenario.

The very high activities of minor actinides, particularly ^{244}Cm, in multiple-recycled fuel for LWR-TRU transmutors may make reprocessing and refabrication so difficult and expensive that transmutation in LWRs would have to be limited to a few recycles. Other studies (Baetsle, 1993) have proposed deploying fast reactors as a means of further destruction of the TRUs after a few recycles in LWRs. In a fast-neutron spectrum the higher mass transplutonic radionuclides will be rapidly depleted.

Thus, one should recognize the need for many fuel-development cycles, entailing extensive in-reactor testing constrained by safety considerations, to build the database necessary for NRC licensing. In addition, the effort entails the refinement and demonstration of isotopic assay techniques and nondestructive fuel testing techniques (e.g., using radiation sources with standard background suppression methods) that can remain effective in the presence of the radioactivity in the refabricated fuel.

For the fuel compositions anticipated during the first few (approximately 30-year) generations of LWR transmutation, the better part of a decade, or perhaps longer, may be required to develop the fuel database, specifications, and manufacturing processes for licensing by the NRC. Assuming that existing fuel fabrication facilities (such as SAF) and existing test reactors could be used, the fuel development, characterization, and in-reactor test program would still require approval by many industry and other participants, as well as governmental organizations. This approval cycle greatly lengthens the time otherwise required to plan and conduct the program. If major new test facilities are required, the additional time to design, approve, fund, and construct them could easily double the estimate of a decade.

An extended period of fuel development, in-reactor test, and demonstration would be needed for the compositions that would occur in the later fuel cycles of the transmutation period. Chapter 4 points out a possible future option—separating the MAs and burning them in a fast reactor should their build-up pose too severe a radiation environment in LWR-fabricated fuel to permit adequate fuel characterization during the later fuel cycles.

Risk And Safety Issues

A major effect of an LWR waste transmuter on repository risk would be to reduce the amounts of long-lived fission products, ^{99}Tc and radioiodine, that tend to dominate the leach-and-migrate scenarios, as well as to reduce the ^{237}Np that would become the dominant long-lived radionuclide once the key fission products were removed. Transmutation also could reduce the TRUs that influence the repository-breach scenarios, such as the plutonium and americium radionuclides. In addition, like other separations and transmutation options, an LWR system could reduce the estimated repository risk by enabling a more optimal wasteform to be used for the residual HLW after transmutation, which must go to a geologic repository along with the ^{90}Sr and ^{137}Cs fission products.

These benefits come at the expense of some level of risk that would be incurred by the operation of LWR waste transmuters and their associated fuel-cycle facilities over the long transmutation period. The preceding subsections note the substantial experience with MOX fuel but point out that burning actinides raises fuel performance and reliability issues that impact reactor safety. Also, the issues relating to control of reactivity and power distribution in the reactor core, discussed in the preceding subsections, would affect the safety evaluation of an LWR transmutation system. Finally, one must recognize the safety issues inherent in transportation of the radioactive materials between sites.

Cost Information

Development Costs

One development cost of significance is the waste form and process to package the residual HLW for geologic disposal. This cost would be similar to that of other transmutation options. For further discussion, see *Impacts of S&T on Waste Repository* , in Chapter 6.

The second significant cost is for the development and performance verification of fuels of changing TRU composition for long burn-up, as discussed in *State of the Technology*. To cover the compositions anticipated over the first several generations of LWR transmuters, the better part of a decade, or perhaps longer, may be required at a cost of perhaps $50 to $100 million per year, exclusive of constructing any major new facilities (E. Evans, private communication, 1993). Part of the testing (e.g., for the later part of the many-decade transmutation period) might be carried out in parallel with operation during the first few fuel cycles in the transmutation mode.

The third significant cost is for TRUEX process development and scale-up through pilot-scale demonstration for aqueous PUREX/TRUEX processing of the LWR spent fuel (see Chapter 3 for details).

System Costs

A recent study of an LWR-based system to burn plutonium from nuclear weapons made a preliminary estimate of initial capital and annual operating costs for an "energy park" comprising up to nine AP-600 advanced LWRs, together with a "fully automated" MOX fuel fabrication plant of 60 to 70 MgHM/yr capacity (Buckner and Parks, 1992). (See discussion above under *Facilities and Support Requirements* .) For each AP-600 reactor, the capital cost is estimated at $1.0 to $1.2 billion. For the MOX fabrication plant, the operating cost is estimated at $30 million/yr, and the capital cost is estimated at $680 to $100 million for uranium oxide conversion, $205 million for plutonium to PuO_2 conversion, and $375 million for pellet fuel production and fuel rod assembly. However, it is not clear that these estimates include waste treatment and an automated materials accountability system. A detailed design of the MOX facility is not yet available. However, its estimated capital cost seems somewhat low compared with that of the SAF line discussed above, which is estimated to cost about $100 million (1980 dollars) for a nominal capacity of 6 MgHM/yr, exclusive of the rest of the fuel-cycle plant, which houses the SAF line.[19]

These estimates for the cost of fabricating military plutonium could be considered a lower bound on the cost of fabricating multiple-recycled MOX fuel, whether from a LWR-plutonium transmutor or a LWR-TRU transmutor. The multiple-recycled MOX fuel will have far greater activities of ^{238}Pu and transplutonic radionuclides and will be more difficult and expensive to fabricate.

Chapter 4 summarizes the impact of fuel fabrication costs on fuel-cycle economics. Recent estimates of the cost of all-remote MOX fuel fabrication, compared with that of LEU fuel, indicate that MOX fuel costs somewhat more than LEU fuel even if the plutonium is available at no cost (OECD, 1992). The differential depends on the prevailing ore and enrichment costs, but is relatively small compared with several other annual costs.

However, the assessment in Chapter 4 concludes that the capital and operating costs of a reprocessing plant for LWR spent fuel make the overall economics of thermal recycle to LWRs strongly negative for the United States relative to a once-through cycle. That chapter estimates the cost of reprocessing LWR spent fuel in the United States at $800 to $1,900/kgHM, depending on the mode of finance and other uncertainties.[20] On that basis, the excess cost would be in the order of $40 to $100 billion for reprocessing the 60,000 MT of LWR spent fuel slated to go to Yucca Mountain and recycling the TRUs to LWRs, rather than using fresh LEU with direct geologic disposal of the spent fuel. This estimate neglects the higher cost of MOX fuel refabrication and the cost saving for reduced uranium ore and enrichment, but these effects are much smaller than the marginal cost of LWR spent-fuel reprocessing. Only at a later time, when the cost of uranium ore has risen to well over $100/lb, would the overall economics of TRU burning in LWRs begin to reach economic parity with using fresh LEU fuel. At such a time, other alternatives that use the fissionable resources more efficiently, such as an ALMR, would have to be considered.

Of course, other societal concerns, goals, and needs might override a mainly economic decision basis for waste transmutation, as discussed in Chapter 4. Nonetheless, the $40 to $100

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[19] The cost of the fabrication line itself, after development and demonstration, was about half of this $100 million estimate. The total includes extensive effort by LANL, ORNL, ANL, the nuclear industry, and other organizations in the United States, ignoring the contributions of the joint international programs and the parallel research and development efforts in other countries (E. Evans, private communication, 1993).

[20] The reprocessing cost range from Chapter 5, $800 to $1,900/kgHM, assumes aqueous-based technology for the oxide LWR spent fuel. That assessment is based primarily on a significant body of data on commercial reprocessing in several large facilities outside the United States using aqueous-based technology, which gives insight into the costs. Recently, ANL has proposed pyroprocessing techniques that might prove suitable for reprocessing LWR spent fuel. The basic chemistry of pyroprocessing has been demonstrated in the laboratory for metal fuels for which ANL is conducting initial scale-up experiments (see Chapter 2). ANL estimates that the pyroprocessing of LWR spent fuel, if feasible, would cost less than aqueous-based reprocessing. Because of the large projected cost saving for LWR spent-fuel reprocessing, the pyroprocessing concepts deserve further development support to determine feasibility and assess the prospects for realizing the projected cost saving.

billion magnitude of the net excess cost for LWR TRU burning is an indication of how serious the situation would have to be before other societal goals might become overriding.

The Los Alamos Accelerator Transmutation Of Waste

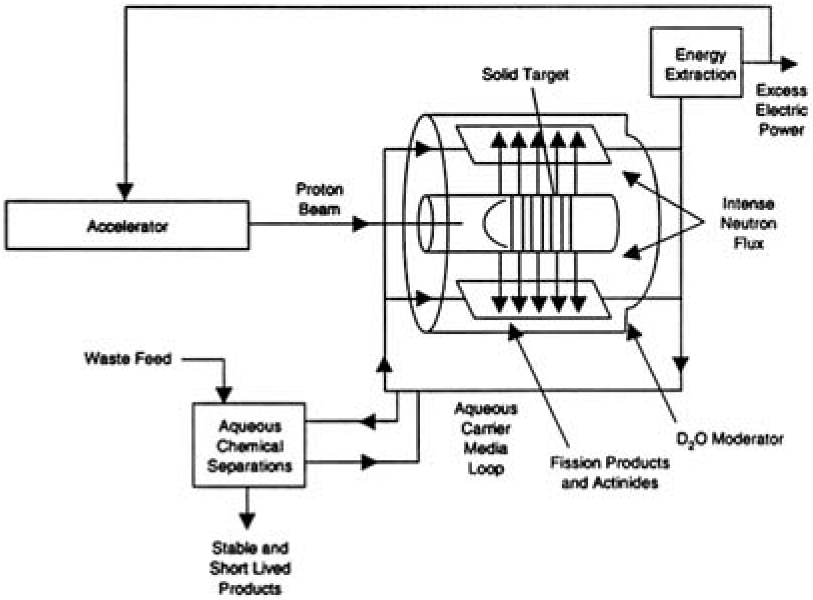
Reference Design for ATW System

The concepts proposed by the Los Alamos National Laboratory (LANL) consist of two primary components, a proton accelerator and a subcritical target assembly. These concepts include high energy protons impinging on a target assembly to produce neutrons. The alleged advantage of a subcritical target driven by an advanced state-of-the-art proton accelerator is in the production of large fluxes of neutrons. The proton accelerator would operate at efficiencies and duty cycles not achieved to date, and the balance of the systems for reactors and reprocessing are also advanced and beyond the state of known art. It should be recognized, however, that LANL has had limited funds to develop these concepts, and the design and concepts presented are undergoing continuous change.

Four concepts for accelerator transmutation of wastes (ATW) are described by LANL: one aqueous heavy-water process and three nonaqueous molten fluoride salt processes. The concepts differ in the type of fuel flow streams, the material used for the target, and whether thorium is used in the molten salt solution fuel to provide production of fissile ^{233}U. The concepts are

1. transmutation of TRUs and some fission products recovered by aqueous reprocessing of LWR spent fuel, using a heavy-water, oxide-slurry aqueous multiplying system that generates excess heat for electric power production and a fast-cycle aqueous reprocessing system (Case ATW-1);
2. transmutation of TRUs and some fission products recovered by nonaqueous chloride volatility or the aqueous reprocessing of LWR spent fuel, using a nonaqueous, molten-salt liquid fuel, graphite-moderated multiplying system that also generates electric power and reprocesses by treating the molten fuel salts (Case ATW-2);
3. transmutation of TRUs and more fission products recovered by nonaqueous chloride volatility or the aqueous reprocessing of LWR spent fuel, which uses a molten-salt, graphite-moderated multiplying system that is fueled partly with thorium, and generates electric power and reprocesses the molten fuel salts (Case ATW-3);
4. transmutation of TRUs and some fission products formed in a nonaqueous self-sustaining thorium-breeder system exclusively for electric power generation and reprocessing in the molten fuel salts for ^{233}U (Case ATW-4).

The ATW system consists of six general components that are schematically represented in Figure F-7. They are an accelerator, a target, a blanket assembly, chemical processing facilities, power production facilities, and waste management facilities. These systems are in the early conceptual phase and will require development of detailed conceptual designs, programs



Source: Los Alamos National Laboratory (1992).

FIGURE F-7 Reference design for commercial waste transmuter.

to develop and test individual components, and prepilot demonstration systems to reach the desired performance levels. Even though the concept for this facility is in an early stage, the existing technology base is sufficiently developed to make some preliminary projections of systems operation and performance.

Performance Requirements

The emphasis in evaluating performance requirements is on the performance LANL expects from the ATW concepts and how they would aid in the disposal of radioactive waste. The clearest understanding of those expectations and how they have evolved as the concepts are refined is obtained by reviewing the goals and objectives set during recent years.

The three relevant principal performance goals that a proposed ATW would be expected to achieve are:

1. transmutation of all long-lived actinide and fission product components of nuclear waste;
2. radiotoxicity of final disposed waste less than that of uranium ore; and
3. reduction of the time scale for nuclear waste storage to that of a human lifetime.

From the discussions with LANL personnel, it was understood by the committee that the goal of the personnel would be to transmute all radionuclides, including those formed in the process of transmutation. While the extent of transmutation has not been fully defined in their materials or reports, they have indicated that their goal is for any remaining waste stream that contains actinides to be suitable for disposal as Class C low-level waste. This suggests that no HLW repository would be needed for these wastes.

While the goal of radiotoxicity less than that of the uranium-ore fuel cycle is an interesting transmutation objective, it is not a particularly useful one in today's regulations. Furthermore, the committee has not seen the basis for such a toxicity calculation, and it could not determine whether the objective is based on curie content of the original ore requirements or curies per unit volume of waste. These issues and the evaluation of waste from the process are discussed in some detail later.

In expecting that the ATW can reduce the time scale for nuclear storage from 10,000 years to a human lifetime, the concepts go further than eliminating high-level waste (HLW). No storage or disposal would be required after the waste has been stored for about 70 years. This means that the longest remaining half-life after transmutation must be only a small fraction of a human lifetime, but details on how this could be achieved have not been provided. During subsequent concept development, LANL stated that no remaining radionuclides would have a half-life exceeding 30 years. Details supporting this are also under development. Presumably, such waste would contain fission products ^{90}Sr and ^{137}Cs (half-life 3 to 10^6 yr) in hazardous quantities. These must be in waste form suitable for emplacement in a geologic repository or monitored surface storage for a duration of at least several hundred years. Understanding of this key issue is important.

The comparative transmutation performance for the ATW is discussed in Chapter 4 where the inventory reduction factor is calculated for the Combustion Engineering pressurized water reactor (PWR) design as a plutonium burner, assuming a process decontamination factor of 1,000. In all cases, the ATWs have even higher reduction factors as a function of years of operation. Similarly, the time required to attain a given inventory reduction would be less for any of the ATW concepts than for an ALMR or a LWR. For example, under a steady power scenario, an inventory reduction factor of 10 requires about 7 years for the ATW-2, 300 years for the LWR, and 430 years for the ALMR. For a reduction factor of 100, it requires about 400 years, 4,000 years, and 7,000 years, respectively.

ATW Concepts

Accelerator

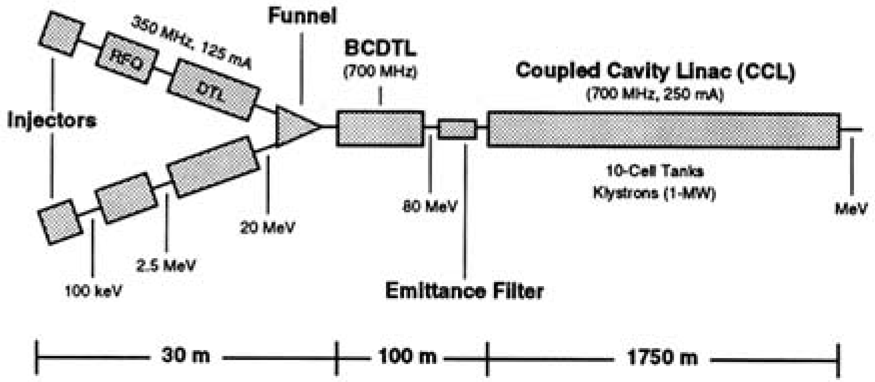
Since the accelerator is fundamental to ATW concepts, it is appropriate to begin the review with the reference design of the two proton accelerators. A general configuration and subsystem description is shown in Figure F-8. The highest energy machine is used to drive ATW-1 and employs a 1.6 GeV, 250 mA (400 MW) accelerator. (Reference LA-UR-92-2020). This machine is proposed to operate in a continuous wave mode with a macroscopic duty factor of 100%. The 1.6 GeV (250 mA) beam is ultimately split into four beams of 62.5 mA each, and these beams impact the target/blanket to produce a neutron flux of about 2×10^{15} neutrons/cm ^{2} sec.

From two injectors, the beams are combined by a funneling structure, then the protons are accelerated from 20 to 80 MeV in a 700 MHz bridge-coupled drift tube linac (DTL) consisting of 86 5-cell tanks with focusing elements utilized between the tanks only. The 700-MHz coupled cavity linac (CCL) accelerates protons from 80 MeV and is made up of 812 10-cell tanks. As in the DTL, focusing elements are only employed between the tanks in order to permit a smoothly changing acceptance and strong focusing.

An important design goal in the high energy portion of the linac is achieving a high radio frequency (RF) efficiency. This is critical, since RF costs dominate both the capital costs of the accelerator and its operating costs. To increase RF efficiency, a low capital cost and high electric conversion factor are necessary (power grid to beam power). A CCL RF-to-beam efficiency of 84% is projected with the current accelerating gradient of 1 MV/m, resulting in an approximately 2-km accelerator.

Other significant design considerations are

- *Proton source*. The proton source for the ATW accelerator requires state-of-the-art performance with respect to output, lifetime, and brightness. The source currently operating at Chalk River most nearly approximates these parameters. It has been operating with a proton output of 105 mA and an RMS emittance of 1.2×10^{-6}-m rad. Only a 10% extrapolation from current performance is required for ATW operating requirements. The emittance is nearly a factor of 2 less than specified for injection into the ATW radio frequency quadrop (RFQ).



	ATW-1	ATW-2, 3, 4
Beam Energy	1600 MeV	800 MeV
Beam Current	250 ma	110 ma
Beam Power	400 MW	88 MW
Total RF Power	475 MW	130 MW
RF to beam efficiency	0.84	0.68
AC to RF efficiency	0.58	0.58
AC power requirement	845 MW	250 MW
Average CCL gradient	1 MV/m	1 MV/m
Transverse output emittance	0.065 π cm - m rad	0.043 π cm - m rad
CCL operative/beam size ratio	11-21	13-25
RFQ Frequency/current	350 MHz, 125 ma	350 MHz, 55 ma
CCL Frequency/current	700 MHz, 250 ma	700 MHz, 110 ma
CCL length/10-cell tank	1750/812	830/550
CCL Klystrons	480	145

SOURCE: Los Alamos National Laboratory (1992)

FIGURE F-8 Accelerator concept for LANL II advanced ATW system.

However, to operate in CW mode, further development is necessary. At Chalk River, a 600-ke V RFQ injected by an ECR source has produced an output of 70 mA in CW operation.

- *RF power sources*. High efficiency RF sources are critical to reducing operating costs. Development of a high-efficiency, low-cost, long-lifetime 700-MHz RF source for the CCL of the ATW would offer the greatest leverage in reducing costs.
- *Beam-loss control measures*. Hands-on maintenance is highly desirable. To allow the hands-on maintenance desired for the ATW accelerator, beam losses must be kept to 1×10^{-5} of total current. Los Alamos Meson Physics Facility operational experience and simulations relevant to ATW have identified areas of beam losses in the CCL. Design features of the ATW linac to minimize losses include a large ratio of accelerator aperture to transverse beam size, a linac front end designed to minimize losses in the DTL, and an emittance filter following the funnel to remove transverse phase space tails before the beam enters the CCL accelerating structures.

  For all other ATW concepts, the accelerator energy and current requirements are reduced to 800 MeV and 110 mA (88 kW). The injectors for the concepts RFQs and DTLs are essentially the same but have lower current requirements. The CCL operates at the same frequency but with fewer 10-cell tanks and accompanying RF power sources.
- *Reliability considerations*. An accelerator availability in excess of 85% would be necessary to meet the needs of a satisfactory ATW system. This has not routinely been achieved with accelerators of this type, which were used in a research rather than a production environment, often undergoing modifications and upgrades to enhance performance in once or two parameters. However, accelerators can be designed with high availability as a specification. There is no component that is inherently unable to reach a resulting goal of 85% accelerator availability with the appropriate R&D effort and a serious preventive maintenance program.

Target System

The target material and design for the ATW concepts vary according to the specific application. Originally, LANL had proposed solid tungsten-lead targets for ATW-1 and liquid lead for the other concepts. Currently they favor liquid lithium metal or molten salt, where there is a reasonable body of experience from work done in support of high energy neutron sources. The spallation targets are designed to yield 30 or more high energy neutrons per GeV of incident proton, resulting in a neutron multiplication factor of between 10 and 20 in a surrounding blanket of fissionable material. With approximately two-thirds of the proton beam energy converted into thermal energy in the target, high power densities of 3 to 4 GWt/m^3 is estimated for the target. This range of power densities is somewhat higher than that of the FFTF but considerably smaller than that of the High Flux Beam Reactor at Brookhaven National Laboratory. LANL analyses show decay heat generation in the target is manageable, with proper heat exchanger designs for rated power operations.

Multiple nuclear reactions involving high energy neutrons in the target produce a host of nuclides, many of which are radioactive. Particular attention has to be given to the containment and management of volatile spallation products (e.g., 10.72-yr ^{85}Kr and 36.4-d

^{127}Xe). In addition, recovery and transmutation of long-lived radionuclides, especially 1.5 × 10^7-yr ^{205}Pb, have to be considered. The equilibrium inventory of ^{205}Pb in a lead target of 3 Mg is estimated to be 15 kg.

A recent target design for the nonaqueous ATW concepts uses a ^{7}Li liquid-metal or molten salt target surrounded by lead or ^{238}U. In this design, the intense proton beam produces a few high energy neutrons in the light-metal target, and the energy of these spallation neutrons is expended in knocking a few more neutrons out of the heavy metal, which essentially serves as a fast neutron multiplier. The lithium flow in the target is connected to the coolant lithium flow in the blanket, which consists of fluid fuel circulating through a graphite block immersed in a lithium pool.

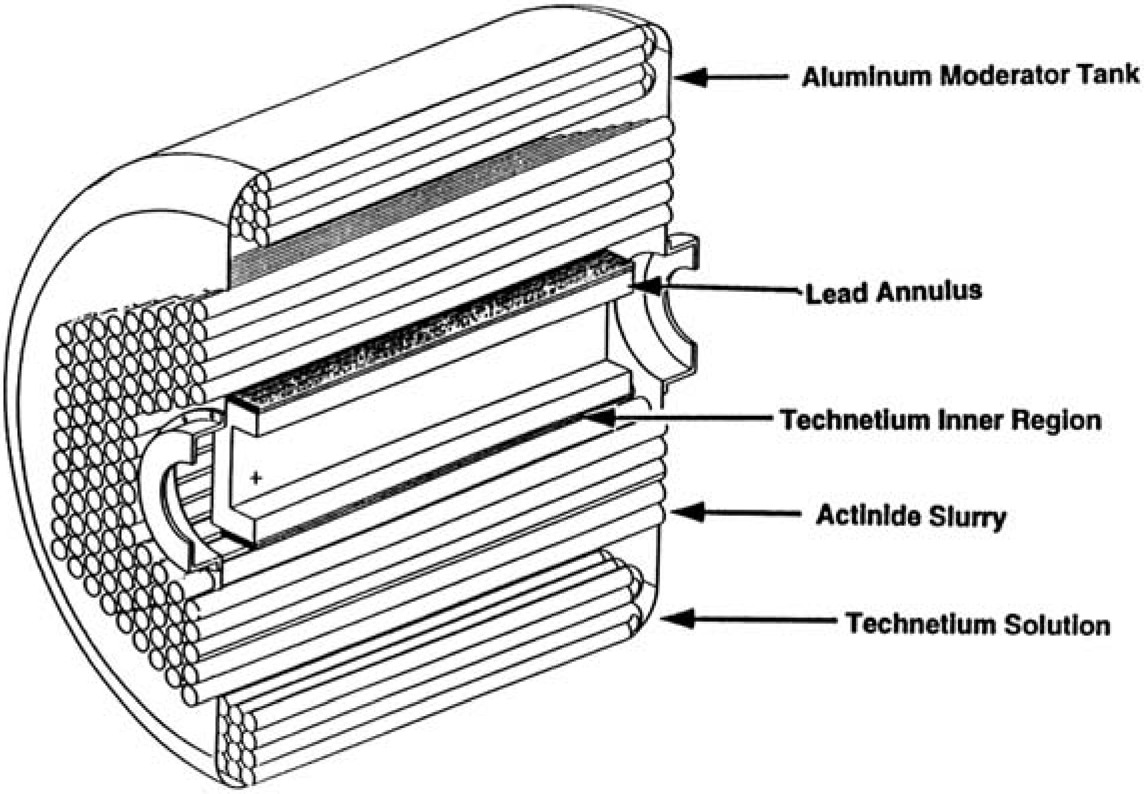
Blanket Assembly

The ATW-1 concept specifies aqueous reprocessing technology to obtain TRUs from LWR spent fuel and to process the blanket and target materials. Separated fission products would be formed into solid targets for transmutation in the thermal neutron flux. The reprocessing systems must provide for high recovery and substantial separation of all TRUs and of all fission products if no high-level and TRU wastes are to be produced. Since the reactor is to be fueled and elements to be transmuted are added with a slurry of oxides and salts in heavy water, with much of the processing postulated in light water, in addition to the complex processing for separations and recovery, there are a number of problems of transition from one solvent to another.

The blanket or reactor structure of the aqueous ATW-1 is patterned after the pressure-tube calandria structure of the Canadian CANDU power reactors. However, the coolant pressure and temperature have been increased to obtain higher thermal efficiencies because of the high electrical energy needed to power the accelerator. Also, there are significant differences in the design of the pressure tubes. Figure F-9 provides a general view of the blanket with different flow streams in the assembly depending on the surrounding neutron flux and energy spectrum.

Chemical Processing

To obtain a separated stream of TRUs and fission products (see Chapter 2), the ATW designs require processing of LWR spent fuel. These TRUs and fission products would be shipped to the site of the ATW subcritical reactors where integrated on-line processing units (1) process the fluid-fuel blanket streams from the reactor core into separate fission products for recycle as special transmutation targets, (2) process the transmutation targets for recovery and recycle, and (3) process the solid or liquid spallation targets to remove and recycle radioactive spallation products. All of these processing steps, including the reprocessing of LWR spent fuel, must have very high recoveries, so that most or all reprocessing waste will qualify for low-level disposal. Some of the ATW integrated processes operate continuously, with minimal cooling



SOURCE: Los Alamos National Laboratory (1992)

FIGURE F-9 Blanket design.

of the streams diverted from the reactor for processing. The processes differ drastically in detail for the aqueous and nonaqueous ATW concepts.

The overall ATW systems model in Figure F-10 illustrates the major differences in the several approaches. Table F-3, which gives the principal design features of ATW transmutation concepts (see below), is useful in comparing these approaches. The critical differences occur not only in the target and blanket assembly but also in the chemical plant and equipment. It is clear that options including thorium or defense wastes as a feed and power production would require somewhat different configurations.

ATW Systems

Having looked at the basic components of an ATW system, and their similarities and differences on a general scale, the specifics of each of these concepts are given below in some detail.

ATW-1

In the ATW-1 design, a 1.6-GeV proton beam produces spallation neutrons that are moderated by heavy water contained at near-atmospheric pressure in a calandria structure. A separate hot heavy-water stream at high pressure, containing a slurry solution of TRUs, fission products, and activation products is circulated through Zircaloy process tubes that penetrate the calandria. The process tubes must be thermally insulated from the heavy water of the moderator. Thermal neutrons must diffuse from the heavy water through the process tubes and into actinide and fission products containing fuel, causing fission and transmutation.

The heavy-water fluid fuel is circulated through an external heat exchanger (where problems with deposits from the saturated slurry-solution fuel working across a high-temperature gradient can be expected) generating steam to drive a turbine generator. A large fraction of the electricity generated is fed back to operate the accelerator. There are four calandria systems, referred to as "blanket modules," coupled neutronically to each other and to the spallation target. Part of the circulating fuel will be delivered to an evaporating system where it will be converted to a dry powder to feed the reprocessing system. The recovered heavy water will be used to make a new slurry from the products of the reprocessing plant.

The heavy-water blankets form a subcritical reactor, operating with an effective reproduction factor, $k_{eff} = 0.95$. This means that 20 times as many neutrons are formed by fission as by accelerator-produced spallation. The subcritical reactor is basically similar to the CANDU heavy-water reactor, except that the CANDU reactor utilizes solid uranium-dioxide fuel in the process tubes and is cooled with heavy water of well-controlled chemistry.

A portion of the heavy-water fluid fuel is circulated to an integrated reprocessing system, where fission products and activation products are removed. This concept anticipates that, with an overall decontamination factor of about 1,000 for a variety of separations and an annual waste volume of 50 cubic meters, the content of TRUs in the resulting waste will be less than

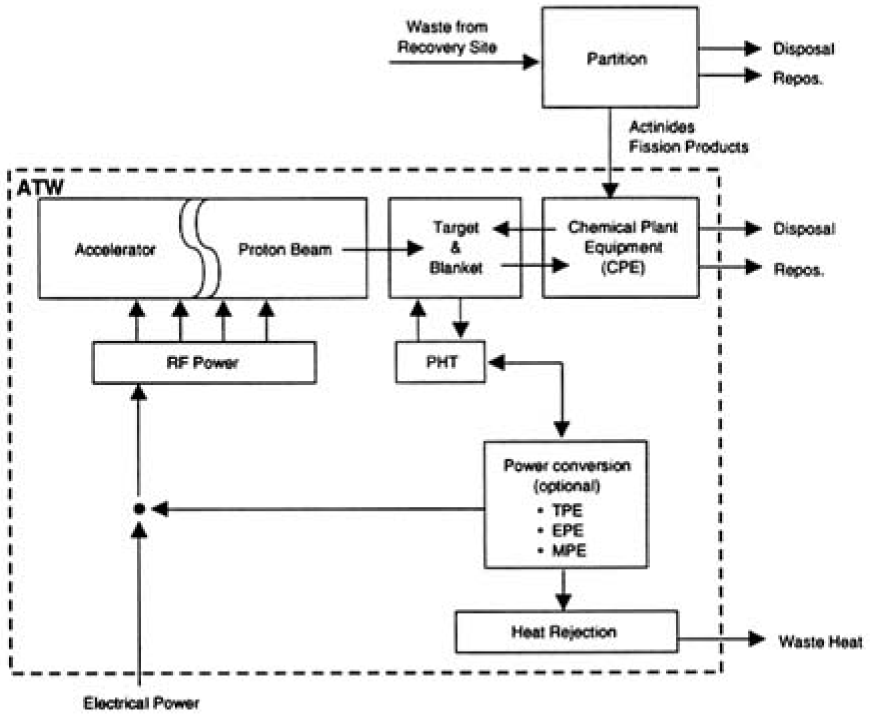


FIGURE F-10 ATW systems model.

TABLE F-3 Principal Design Features of ATW Transmutation Concepts

	Aqueous LWR Fueled	Nonaqueous LWR Fueled	Nonaqueous LWR plus Thorium Fueled	Nonaqueous Thorium Fueled
Thermal power, Mwt[a]	8,3000	6,000	3,000	3,000
Accelerator beam current energy	250 mA; 1,600 MeV	110 mA; 800 MeV	110 mA; 800 MeV	55 mA; 800 MeV
TRU transmuted kg/yr @ 80% capacity[b]	2,602	1,920	240 from LWR	960 from Th
Met electrical power, MWe[a]	1,601	2,180	1,000	1,040
Moderator	Heavy water	Graphite	Graphite	Graphite
Fuel	TRU oxide Slurry in heavy water	TRU in molten fluorides	TRU in molten fluorides	TRU in molten fluorides
Coolant	Fluid fuel	^{7}Li	^{7}Li	^{7}Li
Spallation target	Tungsten, lead	^{7}Li, uranium	^{7}Li, uranium	^{7}Li, uranium
Percent fission products transmuted	3.0	2.5	7.5	7.3
Effective production factor	0.92	0.95	0.95	0.95
Average neutron flux in blanket, n/cm^{2}s	1.3×10^{15}	2×10^{15}	5×10^{15}	5×10^{15}
Cooling system inventory/total TRU inventory	0.296	0	0	0
Processing inventory/total TRU inventory	0.408[c]	0.25	0.25[d] ; 0.75[e]	0.25[d] ; 0.75[e]
Times to process blanket inventory, yr.	0.171[c]	0.083	0.83[f]	0.83[f]
Total TRU inventory, kg	17,850	938	344[d] 806[e]	255[d] 954[e]

[a] The listed thermal and electrical powers are the plant nameplate capacities.

[b] The TRU transmuted per year is the yearly average, calculated at 90% capacity factor.

[c] Two loop averaged values.

[d] Includes rapid chemical separation of protactinium, to decay to ^{233}U.

[e] Protactinium is not separated, 90-day for ^{233}Pa decay before reprocess.

[f] 0.083 day for fission product processing.

SOURCES: Arthur (1992a, b); Bowman (1992a, b); Davidson (1992).

100 nanocuries/g, thereby enabling it to be classified as Class C waste and disposed of as LLW. While the LLW goal for all waste is achievable, it has not been accomplished by any working plant.

The concept specifies aqueous reprocessing technology to obtain TRUs from LWR spent fuel and to process the blanket and target materials. Separated fission products would be formed into solid targets for transmutation in the thermal neutron flux. The reprocessing systems must allow high recovery of all TRUs and of all fission products if no high-level and TRU wastes are to be produced.

According to the concept, a single 8,300-MWe aqueous ATW could transmute the TRUs from eight to ten 1,000-MWe LWRs. The net thermal efficiency of the aqueous ATW is very low, a consequence of the relatively low thermal efficiency of CANDU-like calandria systems and the need to recycle a large fraction of the electric energy produced to power the accelerator. From the data supplied by LANL (Arthur, 1992a, b; Bowman, 1992; Davidson, 1992), calculations show a net thermal efficiency of only 19.3%. Such a low efficiency seriously affects the overall economic performance of the aqueous ATW, as discussed later. The sale of electric energy is a means of mitigating the high cost of chemical processing of LWR fuel or other waste to obtain TRU feed materials for the ATW and the recycle chemical plant for the separations. Other accelerator and reactor concepts have thermal efficiencies of over 30%, which makes their electrical production much more significant.

A material flow diagram for an ATW-1 is shown in Figure F-11, where the material inventories are scaled from the ATW data of Table F-3 for a rated power of 1,000 MWe. The continuous processing of fluid fuel in the ATW-1 concept is represented in terms of equivalent annual material flow rates, with a capacity factor $L = 80\%$ and a reprocessing loss fraction of 0.001. The fuel consists entirely of TRUs, with the inventories in the blanket and heat exchanger and in the reprocessing stream somewhat smaller than the corresponding TRU inventories in typical ALMRs of Chapter 4. The ATW fuel is very rich in MAs, mostly curium isotopes, which make up more than 75% of fuel in the reprocessing stream. Although the overall TRU inventory is relatively small, the discharge and feed flow rates are considerably larger than for the ALMR designs due to a high reprocessing frequency. This results in relatively large TRU losses and achieves rather a small reduction in the overall TRU inventory, as discussed further in this section. The heavy metal depletion rate of 1.63 Mg/yr is high, reflecting a low thermal efficiency eta η of 19% for this aqueous ATW design.

For the aqueous ATW, LANL plans for aqueous reprocessing of LWR spent fuel in some external facility, presumably by an improved, expanded, and complex PUREX/TRUEX-based technology to obtain high recoveries of TRUs and fission products. As shown in Figure F-12, the recovered TRUs are fed into an integrated ATW aqueous reprocessing unit where neptunium and plutonium are separated, converted to oxide slurries, and added to slurry streams returned as recycle fluid fuel to a portion of the reactor core. This portion of the reactor, that irradiates the neptunium/plutonium-rich slurry circulated to an external heat exchanger (which needs to be free of deposits, corrosion, and erosion, even with the CsI in solution) and then to a processing loop is called the "neptunium/plutonium loop." The recycle slurry, which leaves the reactor to be processed, is cooled about 5 days before entering the new and complex

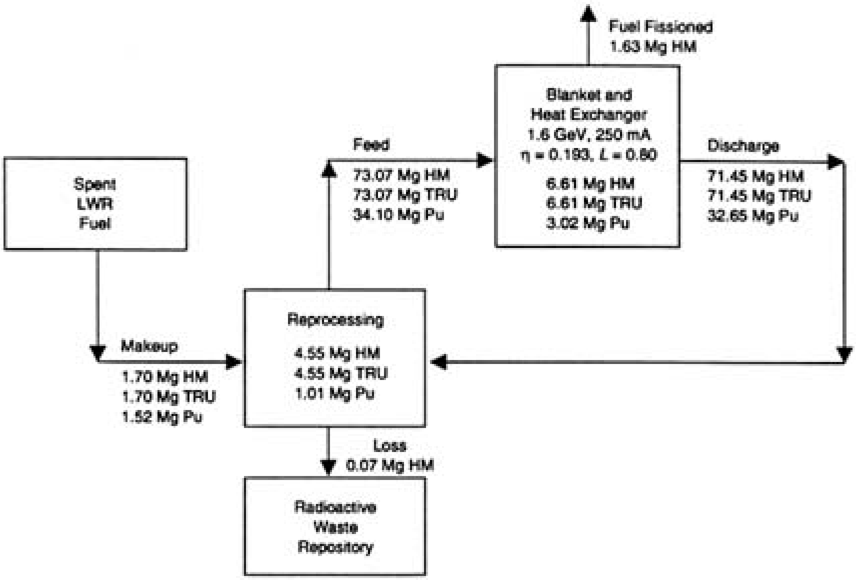
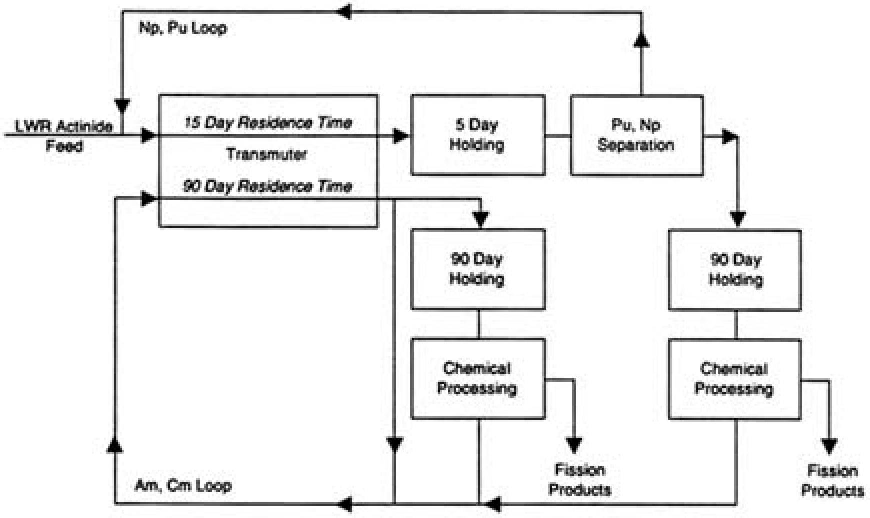


FIGURE F-11 Material flowsheet for one GWe aqueous ATW fueled with spent LWR fuel.



SOURCE: Los Alamos National Laboratory (1992).

FIGURE F-12 Double actinide loop.

high-heat and radiation-load processing unit. The heavy water is removed completely, and the oxides and soluble salts are taken up in a strong acid solution. Difficulty with dissolution is to be expected. The separated plutonium and neptunium are converted back to controlled morphology oxides and reintroduced to the reactor fuel loop. The controlled morphology can be expected to add steps to the process.

Americium, curium, and fission products are separated in the processing unit of the neptunium/plutonium loop and are diverted after holding for 90 days to a separate "americium/curium loop." Americium/curium slurry produced in that loop is circulated through a separate portion of the reactor for irradiation and transmutation and then through another external heat exchanger system with its own fuel inventory and its own heavy-water slurry to light-water-solution transition. These will be slurry/solution loops with individual problems associated with temperature-gradient-driven deposit formation. The slurry stream recycled for processing in this loop is cooled 90 days before processing, with an estimated total external time for reprocessing of 90 days. Neptunium and plutonium formed during irradiation are separated and returned to the neptunium/ plutonium loop, and fission products are separated and sent to the additional fission-product processing system described below.

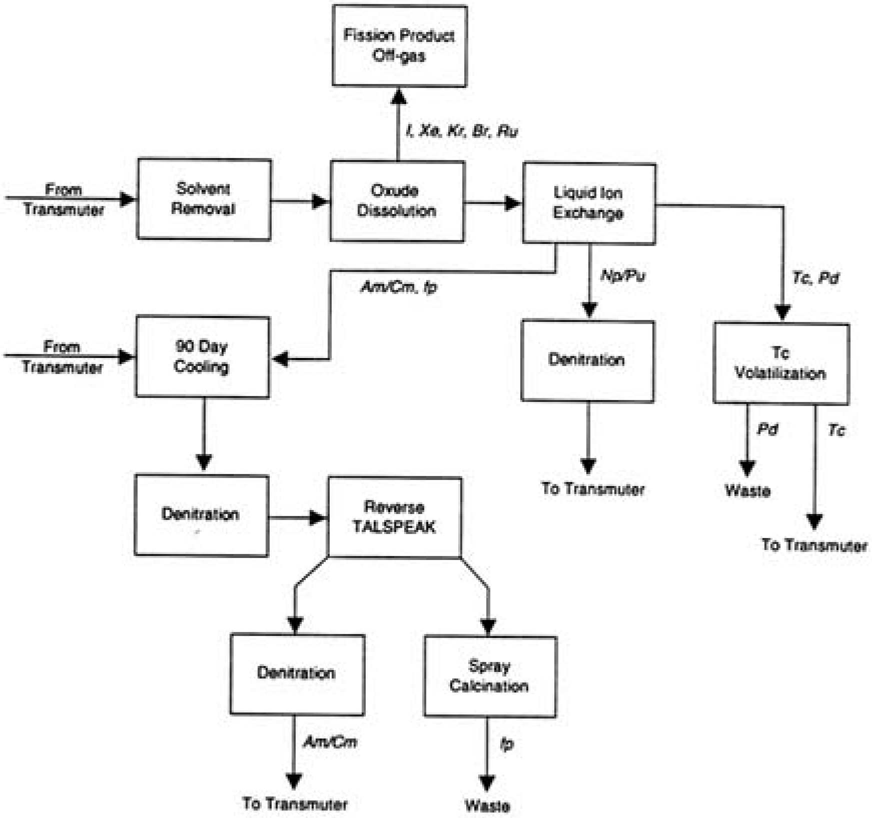
The various integrated processing systems perform separations by aqueous chemistry, as shown in Figure F-13. Incoming feed to the neptunium/plutonium loop is dissolved in nitric acid and pH adjusted. Liquid anion exchange, using a quaternary amine, separates the neptunium/plutonium stream from the stream of americium, curium, and most of the fission products. A separate fission-product stream of technetium and palladium is also produced. The neptunium/plutonium product is denitrated by heating, resulting in oxides that are slurried in heavy water for return to the reactor.

In the americium/curium loop, the separated americium, curium, and most of the fission products, after 90-day cooling, are dissolved, denitrated with formate for pH adjustment, and treated with liquid anion exchange to recover ^{238}Pu that will have been formed by decay of ^{242}Cm. ^{238}Pu is recycled to the americium/plutonium loop.

As shown in Figure F-14, the recovered technetium fission product from internal and external sources is converted to $^{7}LiTcO_4$ in heavy water and fed to the technetium transmuter region of the reactor, which is the third loop that affects reactor operations. Separated fission-product ruthenium is not present in this stream. The ^{99}Tc is converted to stable ^{100}Ru and is discharged by vaporization as RuO_2. Radioactive ruthenium from fission discharges with palladium.

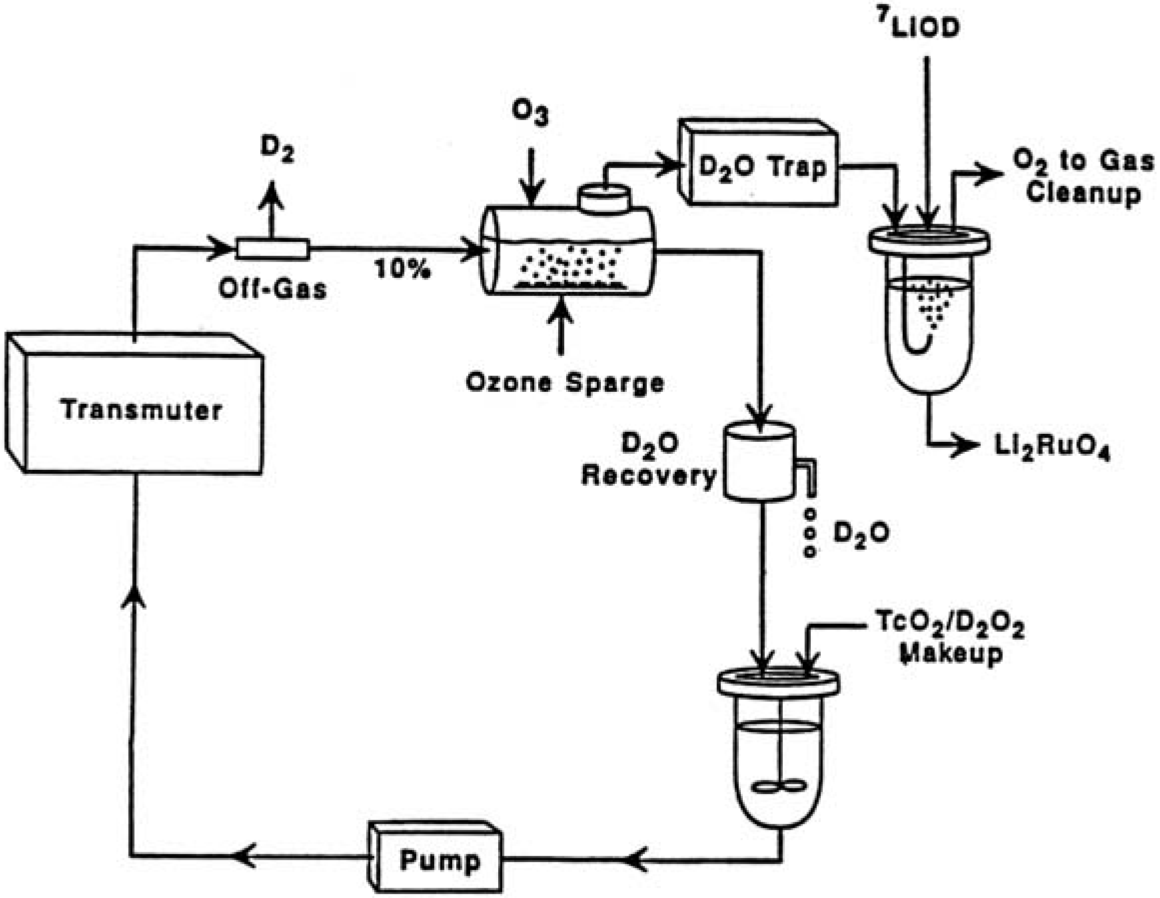
Processes for separating and transmuting fission-product iodine and cesium do not appear in the briefing materials. It has been proposed that the cesium, strontium, and iodine should be collected from the heavy water and the process raffinates and the separated cesium and strontium be allowed to decay in isolation for 10 half-lives. There are no favored processes for these separations as yet. There are suggestions that the iodine should be isolated and exposed for transmutation as the element.

The latest information available to the committee indicates that LANL is giving a low priority to the study of aqueous systems for ATW—molten salt separation process is being preferred.



Source: Los Alamos National Laboratory (1992)

FIGURE F-13 Actinide flowsheet.



SOURCE: Los Alamos National Laboratory (1992)

FIGURE F-14 Ozonolysis option for technetium/ruthenium separation.

Atw 2, 3, And 4

Alternative cases ATW-2, -3, and -4 use the reduced energy accelerator with a ^{7}Li metal or molten salt spallation target cooled by an external heat exchanger. The lithium target is surrounded by heavy metal, lead, or ^{238}U, which multiplies high energy neutrons generated by the incident proton beam. The blanket modules are composed of graphite moderator in the form of machined blocks, fused-salt fluid fuel, and helium coolant. The graphite will have to be of a grade that is a specialty low-porosity product.

Fused-salt fuel is contained in holes machined in the graphite. Lithium coolant flows through other holes in the graphite. Heat from TRU fission is conducted from the molten salt through graphite to the lithium coolant. The lithium coolant flows to external heat exchangers. To reduce production of tritium through thermal capture neutron capture in ^{6}Li, isotopic impurities in the lithium coolant have to be minimized. The ^{7}Li postulated for the molten salts and target is in short supply and not commercially available from U.S. sources. A portion of the fused-salt is withdrawn continuously for nonaqueous processing to remove the fission and activation products with processes not yet demonstrated on any scale. As in the aqueous ATW, no control absorbers have been provided.

From concept data, calculations show the net thermal efficiency of the nonaqueous ATW to be 33 to 36%. Here the temperatures of the fluid fuel and coolant are high enough that an appreciable fraction (39%) of the fission energy is converted to electric energy. Only a few percent of that electric energy is needed to drive the accelerator. The net thermal efficiency of the nonaqueous ATW, as anticipated by LANL, is attractive for generating electric energy for sale to mitigate capital costs and reprocessing costs.

Thorium-fueled modifications of the nonaqueous concept, ATW-3 and ATW-4, are being studied by LANL as a possible means of improving the fuel-cycle economy. Thorium would be added to the fused salt. The concept would generate 75% of its thermal energy from fission of actinides formed by neutron absorption in thorium, roughly equivalent to a breeding ratio of 0.75. The remaining fissions would be from TRUs obtained from LWR spent fuel. Once all LWR fuel has been transmuted, the thorium content could be increased sufficiently for the system to operate as a self-sustaining thermal breeder, analogous to the fused-salt thermal breeder under development at Oak Ridge National Laboratory (ONRL) but set aside many years ago. The advantage projected when the carrier material is molten salt, such as lithium-beryllium-fluoride, is that this configuration can extract electric power efficiently because of the salt's high thermal-to-electric conversion capabilities. Figures F-15, F-16 and F-17 provide materials balance for 1,000-MWe concepts ATW-2, -3, and -4, respectively.

In contrast to the aqueous ATW-1 concept, where the fluid fuel serves as coolant dissipating heat in the heat exchanger, coolant in all of the nonaqueous ATW concepts is liquid lithium flowing through the graphite moderator. Elimination of the fuel inventory tied up in the heat exchanger, coupled with a high thermal efficiency (e η = 33-36%) and higher neutron fluxes, results in considerably smaller TRU inventories for the nonaqueous concepts summarized in Figures F-15, F-16, and F-17. The ATW-2 fuel consists entirely of TRUs from spent LWR fuel and hence shows the largest fractional TRU depletion rate of all ATW concepts. For ATW-3 and ATW-4, either partially or fully fueled with thorium, the materials balance includes

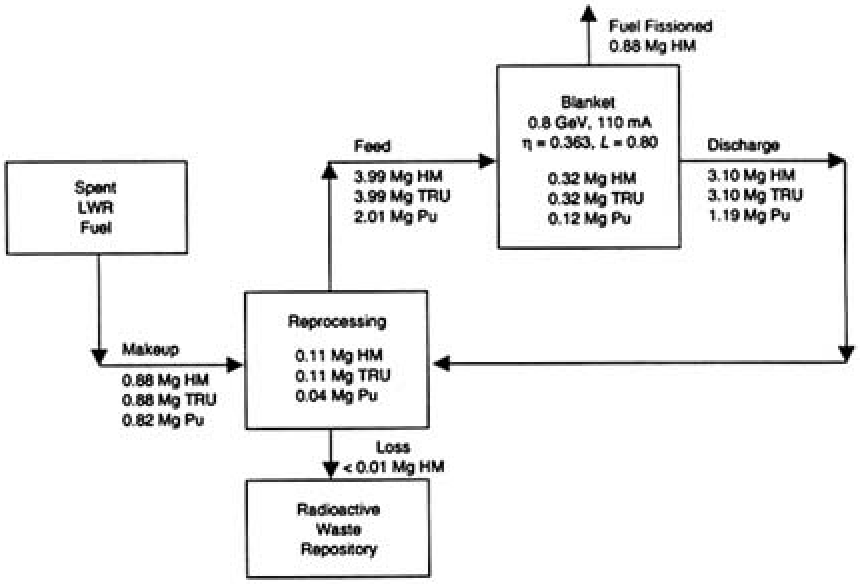


FIGURE F-15 Material flowsheet for 1.0-GWe nonaqueous ATW fueled with spent LWR fuel.

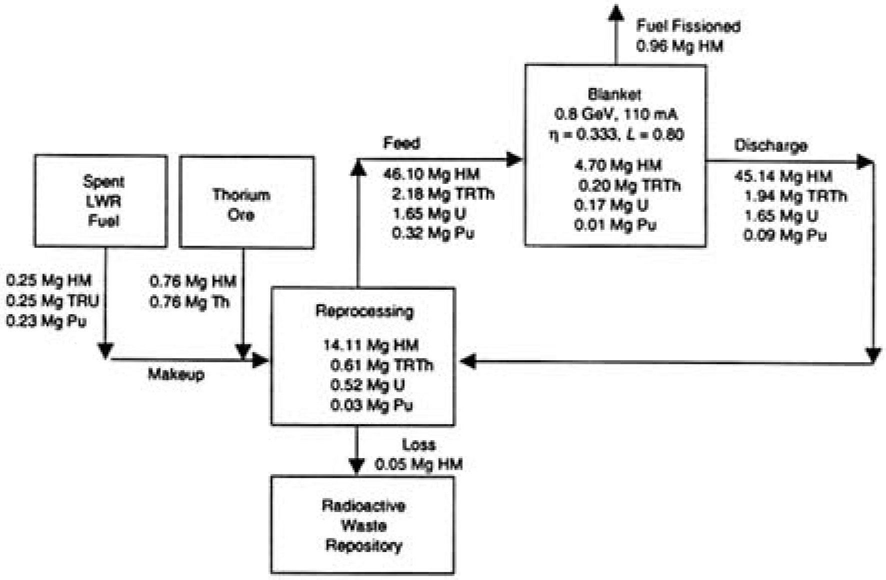


FIGURE F-16 Material flowsheet for 1.0-GWe nonaqueous ATW fueled with thorium and spent LWR fuel.

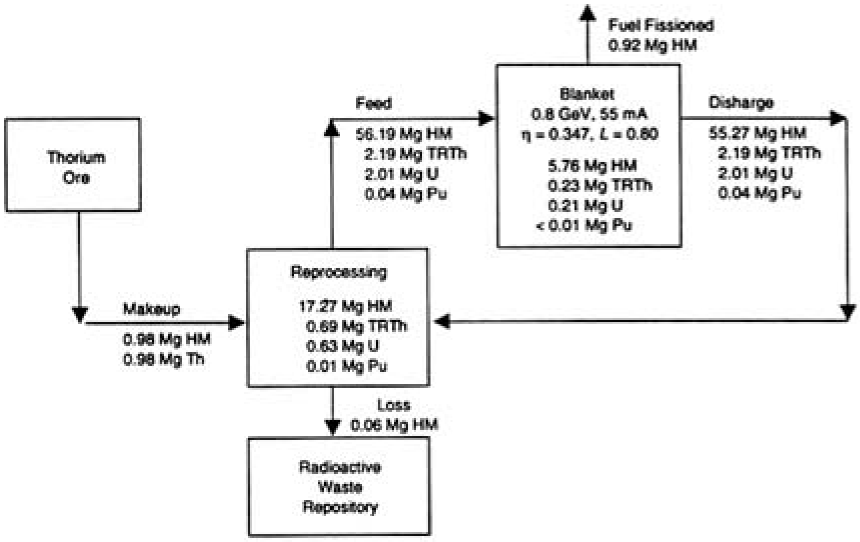


FIGURE F-17 Material flowsheet for 1.0-GWe nonaqueous ATW fueled with thorium.

trans-thorium inventories consisting mostly of protactinium and uranium isotopes produced in the blanket. The flowsheets are limited to the concepts that do not allow rapid chemical separation of ^{233}Pa from thorium to decay to ^{233}U. When rapid separation of ^{233}Pa is allowed, the trans-thorium inventories are smaller, as shown in Table F-3. Although the TRU inventories, mostly consisting of plutonium, are rather small for the ATW-3 and ATW-4 concepts, the actinide transmutation efficiency should be evaluated on the basis of the total trans-thorium inventories for these concepts.

Nonaqueous Reprocessing Of Law Spent Fuel

For the nonaqueous ATW, fueled with a solution of TRUs in molten fluorides, LANL describes entirely different separation processes. The separation of TRUs and fission products from LWR spent fuel is to be carried out in a separate facility, as in the case of the aqueous ATW. However, LANL proposes that the LWR spent-fuel recovery will not be based on the well-established PUREX technology, supplemented with TRUEX for high recovery. Instead, it proposes a water-free halide volatility process, based on technology that has been developed for reprocessing metallic uranium-zirconium alloy naval fuel at Idaho Falls. The process was proposed by Westinghouse (McLaughlin, 1992). It is said to accomplish the required conversions and separations with a minimum of unit operations and a minimum of new technology developments. LANL has not sufficiently explained the desirability of developing such a process for low-enrichment LWR oxide fuel rather than adapting the conventional aqueous process.

The proposed process would remove Zircaloy cladding by adapting a process using a plasma torch. Zircaloy cladding would be reacted selectively with chlorine gas, forming volatile zirconium tetrachloride. The zirconium tetrachloride would then be condensed, separating it from the noble fission gases krypton and xenon.

It appears that the recovered zirconium tetrachloride would have to be sent for geologic disposal. It would be normally expected that this very reactive material would be converted to a more stable form like the oxide for disposal. The generation of hydrochloric acid from a damaged container of Zr ^{14}C in the repository could be a serious problem. The fate of the important radionuclides ^{129}I, ^{131}I, and ^{14}C is not mentioned. At least some of the iodine would be expected to be found in the zirconium chloride. There is appreciable ^{14}C in the Zircaloy, and much of the noble fission gases and iodine will evolve when the cladding is removed. The volume of zirconium tetrachloride waste is likely to be much greater than the volume of compressed cladding hulls resulting from commercial aqueous processing. Its very high reactivity with water and its content of fission products and actinides will present problems.

The remaining solids, containing the uranium dioxide, fission-product oxides, and TRU oxides would then be fluorinated with hydrogen fluoride (HF) to form uranium tetrafluoride, and other volatile fluorides, and then with fluorine to form volatile uranium hexafluoride and other fluorides. Volatile fluorides of TRUs and some fission products like neptunium, plutonium, and technetium will also evolve. A high-purity uranium product stream is said to be obtained by multistage distillation of the gaseous fluorides. Processes of this nature have been used to prepare uranium hexafluoride for the diffusion plants with the creation of by-product streams and have left some technitium, neptunium, and plutonium in the fluoride. The remaining fluorides of TRUs and fission products form the feed to the ATW molten salt blanket.

Whether the proposed process could achieve the high recoveries of actinides and fission products necessary to accomplish the LANL system objectives has not been discussed. It seems that LANL may favor this process, because the ATW blanket is already intended to be a molten fluoride system. However, the alternative of using the well-established existing PUREX technology, supplemented with TRUEX, for LWR spent-fuel reprocessing warrants consideration. There are substantial problems: (1) added salts to waste with the processing of the mixed fluorides in aqueous systems, (2) recovery of the costly beryllium and ^{7}Li; and (3) control of corrosion, complexing, and precipitation problems introduced by the fluoride ions in nitrate chemistry processes.

The idea of chemically decladding Zircaloy-clad UO_2 fuel is not new. A process called the Zirflex process was proposed in the early 1960s before the mechanical decladding was fully developed. It involved dissolution of Zircaloy cladding as ammonium fluorozirconate in a boiling solution of ammonium fluoride containing ammonium nitrate (Smith, 1960). However, mechanical decladding of LWR spent fuel has been proven successful in current commercial reprocessing operations in France, the United Kingdom, and Japan.

There are good reasons why such a process may be attractive for low TRU, low ^{238}U zirconium-uranium alloy spent fuel from naval reactors. However, it is difficult to see how this process could be construed to require less development than conventional aqueous processing when applied to low-enrichment oxide fuel. Also, it is not clear why the proposed process offers any economic incentive over aqueous processing, even if it were fully developed.

Whether the proposed process could achieve the high recoveries of actinides and fission products necessary to accomplish the LANL system objectives has not been discussed. It seems that LANL may favor this process because the ATW blanket is already intended to be a molten fluoride system. However, the alternative of using the well-established existing PUREX technology, supplemented with TRUEX, for LWR spent-fuel reprocessing warrants consideration.

Reprocessing At The Nonaqueous Atw

LANL (Bowman, 1992) proposes that integrated processing for the nonaqueous ATW be based on ultracentrifuge separation of the TRUs and fission products from the BeF_{2-} LiF fused-salt carrier. This novel concept would involve separation of solutes of different molecular weight rather than separation of materials initially present in a two-phase mixture with different densities of phases. Recycle streams and centrifuge angular velocities are adjusted to yield a separated stream of fission-product fluorides. The calculated decontamination factor for removing actinides from fission products is 10^4 to 10^7. The TRUs and remaining salt are recycled.

Centrifuges can be used if reliable machines, remotely maintained, can be created for high-temperature operations. There are problems with the phase diagrams of the systems at hand. Serious deviation from the feed composition will change the melting temperature to higher values on at least one end of the separation in the system, and eventually the problem is one of dealing with the removal of small amounts of impurities from high melting fluorides with melting temperatures to about 1,000° C. The pure liquid phases have to be above their melting points to obtain the high decontamination factors and discharge concentrated wastes.

The light lithium fluoride melts at 848° C and the eutectic with beryllium fluoride at about 350° C at about 50 mole percent. The phase diagram gives a useful liquid range at about 450° C, and fortunately the beryllium-lithium pair, with a mass difference of 2, is about as hard to separate by centrifuge as the lithium isotopes. The lithium and beryllium will tend to separate from each other a little, but the whole separation could be done at the lower temperature using dilute melts of the elements to be separated, refeeding of the isolated light lithium, beryllium fractions with no effort to isolate pure heavy elements in high concentrations with the centrifuges. In phase diagram terms, the isotope separations are the easy ones for simple melts like CsCl. These problems are much the same as those encountered in the separations performed in many stage thermal diffusion columns, such as those used to do some enrichment of the isotopes of uranium in liquid uranium hexafluoride at ORNL in 1944.

LANL also proposes a similar cascade of centrifuges to remove the spallation products from the liquid-lead spallation target. Another cascade would separate the ^{137}Cs isotope from fission-product cesium liquid metal. In the absence of stable ^{133}Cs that could absorb neutrons to form more ^{137}Cs, the ^{137}Cs would then be separately irradiated and transmuted to stable barium. Evidently LANL's purpose is to reduce the content of ^{137}Cs so that the remaining cesium would reach Class A levels for radioactive waste. LANL has not addressed the longer-range problem of ^{135}Cs. Because of its long half-life of 2.3 million years, ^{135} Cs is frequently calculated to be one of the most troublesome radionuclides in geologic disposal of HLW. It should present a similar problem for LLW.

LANL has not presented any extended comments on how this centrifugal separation will be accomplished in molten salts. There have been no commercial applications of high-temperature centrifugation except for some centrifugal casting of metals —rather simple, short-term applications. The applications suggested would require very long periods of continuous operations with very difficult maintenance problems essentially eliminated. The separations proposed with molten salts will lead to phase separations as solids unless there is extensive knowledge of the phase diagrams involved or the operations are conducted above the melting points of the highest melting phase expected. This approaches operating temperatures of a 1,000° C for high thorium fluoride systems. The ability of centrifuges to separate in homogeneous phases is greatly reduced at higher temperatures. This higher temperature requires higher and higher speeds and thus higher stresses with materials of construction that become weaker and weaker for a given separation. The gas machines have a very low mass inventory that allows good use of the mass to generate the needed flow pattern at the expense of even more stress. The only material known to be useful under these expected extreme conditions would be the very expensive, very difficult to make composites of carbon fibers held together with carbon deposits. Billions have been spent on the development to commercial usefulness of the relatively simple and well-known gas centrifuges. The development work has been aimed at operating the centrifuges at near-ambient temperatures with materials that are easier to use and that have efficient isotope separation properties and appreciable vapor pressures at operating temperatures. A comparable expenditure would be expected to be needed for very hot liquid machines if they were possible at all.

Power Production Facilities

The power production part of this concept in heavy-water form is reasonably conventional and based on the CANDU reactor systems. Heat exchangers are used to heat a working fluid, which is used to power turbine generators. The efficiencies of the various approaches are quite different depending on the pressures and temperatures of the system. These efficiencies are discussed in some detail later. The molten salt designs certainly have the potential of being efficient power producers.

Waste Management Facilities

It is the original intent of the ATW concept that only LLW would be produced as a consequence of this program. Several questions have been raised in previous sections concerning the viability of this goal, but not its cost or consequences. A better perspective is provided on this goal in *Performance Requirements* above as well as in discussions under *State of the Technology*.

Safety Issues and Reactivity Control

LANL states that the main benefit from the accelerator would be to allow use of a subcritical reactor for neutron multiplication, thereby avoiding the criticality safety issues of critical reactors and possibly the public issues on nuclear power. Clearly, in this concept neutron multiplication can be stopped quickly by terminating the beam current impinging on the spallation target. The accelerator neutron source would be a rapidly acting control element and should be capable of controlling those reactivity transients that lie within the control capability. For example, if the subcritical blanket systems operate at an effective reproduction factor of 0.95, the accelerator neutron source would control a positive reactivity swing of no more than 5%. LANL does not propose control absorbers for additional reactivity control. Possibilities of reactivity swings exceeding 5% are discussed later.

In the following sections, a number of safety and reactor control issues are discussed. And while this is a subcritical assembly, there are many issues related to control and safety that need discussion. The discussion starts with the accelerator and moves to some materials issues of the target and then emergency cooling, reactivity excursions, fluid boundary-layer heating, pressure-tube failure, and the possibility of explosive gases released from radiolytic decomposition.

Accelerator Safety

The specifications for the ATW accelerator require that significant attention be given to safety considerations such as electrical power, radiation, safety control systems, safety procedures, and training of staff. The intensity of the beam makes it very important that appropriate fast shut-down and interlock systems be in place to protect both hardware and personnel. Although the experience at LANL should provide a solid technological and procedural basis for dealing with these accelerator safety issues, the beam power required necessitates careful scrutiny scaling of these issues.

The alleged advantage of the ATW system over a reactor system is that beam generation can be halted within small fractions of a second. However, radiation is still an issue in the form of neutrons from operation of the beam and beta and gamma radiation from handling, storage, and transport of activated components. Both forms of radiation will require proper shielding to protect health, safety, and the environment. With hands-on maintenance as a design goal of the ATW concept and the experience base at LANL, a database should be created that will allow these issues to be addressed. Mobile remote handling equipment should be available to deal with activated components that need to be removed.

In order to have a safe accelerator, projected beam losses need to be established early in the design phase to permit appropriate choices of materials or provisions for remote handling capabilities. Since a high average availability needs to be achieved, critical components need to be placed in an environment that allows access during running. For those components that cannot be moved behind a shielding wall, modern designs and quick-replacement schemes need to be developed. The emittance filter proposed by LANL needs careful design to permit the stopping of unwanted beam of the halo before it reaches the area where remote handling is available. In addition, interpretation of "as low as reasonably achievable" criteria needs to be carefully considered in the design.

Target Materials Issues

The targets for the ATWs are mixed high and low atomic number (Z) materials that produce large numbers of neutrons per proton, because the ATW systems require high (thermal) neutron fluxes to effectively fission some of the MAs before they decay to nonfissile forms, the power densities in the targets will be large, and for economic reasons the target must withstand high proton/neutron fluences. The proton energies are large (800 MeV and 1,600 MeV), and hence the neutron flux spectrum will include a high energy tail (i.e., a population of neutrons with E>20 MeV). This is uncharacteristic of fission reactor spectra, where most of the irradiation damage experience has been obtained, and it is important, because in addition to displacement damage associated with the high energy protons and neutrons, there could be a substantial transmutation rate associated with endothermic reactions at energies above threshold energies in the MeV range. Transmutants such as helium gas, which can dramatically alter mechanical properties of materials of construction (ductility, creep rupture life) and dimensional stability (creep and void swelling), fall into this class. A host of other transmutants, for which the effects are not as well known, can be produced in significant quantities as well.

Emergency Cooling

The principal safety issues of power reactors arise from fission-product heating after shutdown. The problems of shutdown cooling, to avoid overheating the fuel, coolant, and moderator, are more severe the higher the operating power density. Therefore, safe and adequate shutdown cooling of the ATW is particularly important. An interesting feature is the large proportion of the working fuel inventory that is outside of the reactor at any given time. The power density in the fissioning blanket of the proposed aqueous ATW averages about 250% greater than in a pressurized LWR, so even more severe problems of shutdown cooling are anticipated. In a LWR the main function of the emergency cooling system is to remove fission-product decay heat rapidly enough that the solid fuel does not overheat and release fission products to the coolant. Fuel melting is to be avoided, so containment of fission products by intact solid fuel can be relied on except in extremely improbable degraded-core accidents. The high power-density problems with solid fuels are largely eliminated with these liquid fuels. At the hot end of the heat exchangers, the high power density is expected to cause problems with slurry and solutions condition changes and the erosion and corrosion of the reactor tubes.

In the ATW, fission products, as well as all of the actinides, are already in the coolant, so they would be transported more easily beyond the primary containment in the event of a failure in the containment system. Therefore, there must necessarily be an even greater premium on primary containment integrity for the fuel and blanket both inside and outside of the reactor vessel in an aqueous or molten salt ATW than in fuel rods for LWRs.

LANL proposes to solve the problems of shutdown cooling by draining the fluid fuel from the reactor. However, no details of a fluid-fuel drainage system sufficiently rapid to overcome the loss of the pressurized fluid fuel through a breach in the primary containment have been described. Further, draining the fluid fuel would, in itself, not be sufficient. The fluid fuel would still be generating decay heat at a high volumetric rate. Following the precedents for emergency cooling systems required for licensed solid-fuel reactors, the ATW fluid fuels must be promptly and adequately cooled in order to (1) prevent overheating that would result in high pressures, (2) prevent boiling, and (3) prevent volatilization of many of the radioactive species into an even more mobile form for potential transport from the primary containment. A substantial part of the activity in the reactor fuel cycle will be in solution, and the majority will be in a slurry that can settle to increase local energy densities.

Reactor licensing in the United States and in most other countries requires safety systems to ensure containment of fission products in the postulated event of a sudden break of a main coolant pipe. However, in the ATW fluid-fuel reactor concepts, there are pipes continuously carrying highly active fluid fuel that contain actinides and fission products from the reactor vessel. In the aqueous ATW the fluid fuel is itself the main coolant as it flows from the reactor to an external heat exchanger. It would appear that none of the ATW concepts could survive the present pipe-break criterion.

Although continuous processing of the ATW fluid fuel would remove some of the heat-generating fission products from the fluid fuel, those removed would have half-lives on the order of the blanket residence time of a month or longer. It is the fission products of much shorter half-life that contribute most significantly to the need for a promptly acting shutdown cooling system. The processing rates described by LANL would not be sufficiently rapid to mitigate the most severe shutdown heating transient with assumed loss of normal cooling, which typically occurs very soon after reactor shutdown with loss of normal cooling. LANL has not described how it expects to effect emergency shutdown cooling of the ATW concepts.

Reactivity Excursions

A subcritical reactor is not exempt from problems of reactivity excursion. Fluid-fuel reactors typically have very large negative temperature coefficients of reactivity, because the fluid fuel expands more with heating than does the solid structure of a heavy-water calandria or of the graphite moderator. Therefore, a large reactivity swing can be expected in going from a hot operating condition to cold shutdown. It may exceed the control capability of the accelerator. LANL has not described how it plans to control this reactivity swing with four flowing loops, depending on rate and content in the heavy-water system that are involved with the neutrons. Fast-acting solid control absorbers may be required for scram and safe cold shutdown.

Another issue that is key to the LWR community has to do with fuel characterization and fuel homogeneity. In the slurry-fed ATW burning actinides, actinide thorium mixtures, or even thorium by itself, a standard problem is reactivity homogeneity as the fuel is pumped through the assembly. There are concerns that reactivity transients mentioned earlier can be a continuing issue and that accelerator current control will not be adequate by itself to provide assured subcritical operation.

The ATW concepts would be particularly vulnerable to xenon oscillations. Thermal reactors with sufficiently low neutron leakage and sufficiently high neutron flux tend to undergo large spatial oscillations in neutron flux and power density, even if the total power is held constant. The oscillations appear in the form of traveling waves of neutron flux and power density, usually traveling azimuthally around a cylindrical reactor core. Local overheating of the fuel and structure can result. The higher the neutron flux, the sooner the onset and greater the magnitude of xenon oscillations.

Xenon oscillations are a significant safety issue in commercial LWRs and especially in the heavy-water production reactors, such as those at Savannah River. LANL has provided no information concerning the potential problems of xenon oscillations in the ATW. The reactor structure would have to be heavily instrumented to detect the onset of xenon oscillations. Traditionally, xenon oscillations are controlled by carefully programming the spatial insertion of control absorbers to dampen the xenon peaks as they occur. Many control absorbers are required and must be adjusted spatially as the traveling wave tends to form. It is not clear that the accelerator, with four fixed target locations, is capable of controlling xenon oscillations, other than by turning off the reactor. To be effective, control absorbers must absorb some neutrons even during steady-state operation. Therefore, some reduction in the neutron economy claimed for the ATW can be expected.

Because xenon is not highly soluble in fluids, it is possible that xenon gas may escape from the fluid fuel rapidly enough to suppress xenon oscillations. The aqueous ATW may perform better in this regard, because large amounts of a stoichiometric mixture of deuterium and oxygen are expected to be produced from radiolytically decomposed heavy water that carries the suspension of fuel particles. Stable slurries usually require small particle sizes, and the small fuel particles will allow a significant fraction of fission fragments to escape into the heavy water that carries the fuel slurry, resulting in radiolytic production of a stoichiometric mixture of deuterium and oxygen and the formation of solutions and colloidal dispersion of fission products. The deuterium and oxygen must be removed rapidly to avoid gas pocketing and explosive mixtures. This could sweep out much of the xenon. The turbulence resulting from rapid flow of the fuel slurry to the external heat exchanger may also aid xenon removal. However, no details of the method and rate of removing radiolysis gases and xenon have been provided.

Xenon oscillations may pose a more severe problem for the nonaqueous fused-salt fluid-fuel ATW. No large quantities of radiolysis gas are expected in the fused salt, and the fused salt is not rapidly circulated within the reactor or to an external heat exchanger. It would be more difficult for xenon bubbles to nucleate and escape. Also, xenon can penetrate the graphite moderator that is in contact with the fused salt, thereby decreasing its mobility to escape from the reactor core. Xenon intrusion from molten-salt fuel into graphite was observed in the molten-salt reactor experiment that operated for several months in the 1960s (McNeese and Rosenthal, 1974). It introduced a problem of reactivity control, although the neutron flux was too low and the neutron leakage too large for the onset of xenon oscillations. It was concluded then that graphite in contact with molten-salt fuel must have sufficiently low permeability to gases to limit absorption of Xe^{135} into the graphite. Some laboratory experiments on sealing the graphite surfaces with pyrolytic carbon showed low permeability, but the large increase of permeability under neutron irradiation was a concern. The program was canceled before a successful sealing technique could be demonstrated. Therefore, xenon oscillations in the nonaqueous ATW remain a significant safety issue.

A reactivity problem unique to high-flux thermal reactors is the large reactivity transient associated with the build-up of the 53.1-hours ^{149}Pm, a fission product that decays to stable ^{149}Sm, which has a very large absorption cross section for thermal neutrons. During reactor operation, ^{149}Pm builds up to a steady-state inventory, such that its rate of decay equals its rate of production. ^{149}Sm also reaches a steady-state inventory, such that its rate of neutron absorption equals its rate of production by ^{149}Pm decay. After reactor shutdown, or even after a reduction in power level, the ^{149}Pm decays and the ^{149}Sm builds up to a higher inventory than existed during operation. If the reactor has been operating at sufficiently high neutron flux, the build-up of ^{149}Sm after shutdown is so extensive that it is impossible to start the reactor again. Even a reduction in power level can cause sufficient build-up of ^{149}Sm to shut down the reactor. For a high-flux thermal solid-fuel reactor, the only solution is to unload enough fuel and replace it with new fresh fuel so that the reactor can be restarted.

Here the fluid-fuel reactor coupled to integrated chemical reprocessing offers the promise of eventual restart by selectively removing the ^{149}Sm chemically. However, the processing rates described in the ATW publications would seem to be too slow to prevent the formation of large concentrations of ^{149}Sm, which approaches its peak concentration after shutdown with a time constant determined by the half-life of ^{149}Sm (53.1 hours). Therefore, loss of an ATW reactor core for very long periods can be expected from power fluctuations that can initiate the growth transient of ^{149}Sm. LANL has not indicated whether its ATW concepts would be capable of handling these large ^{149}Sm transients. The addition of reactivity to the circulating slurry or molten salt is expected to be a rather straightforward feature. The short turnaround cycle for the fuel in the heavy-water reactor system is designed to isolate the rare earths, such as samaricium, quickly.

The nonaqueous ATW fueled partly or completely with thorium will be subject to another reactivity transient that is a potential safety issue at high neutron flux. Fissile U^{233} is generated by neutron absorption in ^{232}Th, forming ^{233}Th (half-life 22.3 minutes), and then ^{233}Pa (27.4 days) that decays to ^{233}U. ^{233}Th, ^{233}Pa, and ^{233}U reach equilibrium concentrations at steady state. The equilibrium concentration of ^{233}Th and ^{233}Pa increases with increasing neutron flux. After operating at steady state, a reduction in power level will cause the inventory of ^{233}Pa to increase above the new equilibrium value for a while, followed by a transient increase in the inventory of ^{233}U and an increase in reactivity. If the reactor has been operating at a sufficiently high neutron flux, the reactivity excursion caused by the ^{233}Pa transient can make it difficult or impossible to shut the reactor down. Even a reduction in power can create a reactivity excursion that must be controlled. The LANL ATW designs contain no control absorbers for protection against such reactivity excursions. It is, of course, possible that sufficiently rapid on-line reprocessing of the fused-salt fuel and preferential removal of ^{233}Pa could prevent the positive reactivity excursion characteristic of thorium-fueled reactors operating at high neutron flux. Processing of fluid fuel to separate ^{233}Pa every 2 hours is considered as an option for ATW-3 and ATW-4 and may help alleviate concerns over the ^{233}Pa–^{233}U transient.

Fluid-Fuel Boundary-Layer Heating

New safety issues arise from the radical core designs of fluid-fuel reactors. For example, a reactor in which the fluid fuel is circulated from the reactor core to an external heat exchanger for heat removal is vulnerable to boundary-layer heating of a flowing fissioning fluid fuel. As the fuel slurry flows through a Zircaloy pressure tube in the aqueous ATW, the fluid near the inner surface of the pressure tube flows at a local velocity that approaches zero near the pressure-tube surface. However, the neutron flux is large near the surface, even larger than near the center of the pressure tube. The slower-moving fluid in the boundary layer reaches a higher temperature than the bulk fluid, high enough that heat transported from the boundary layer into the rapidly moving bulk fluid makes up for the lack of convective heat extraction in the slowly moving boundary layer. The higher local temperatures at the pressure-tube surface weaken the pressure tube, are likely to increase corrosion, and may result in local boiling of the fluid fuel.

Fissioning boundary-layer heating contributed to the failure of the inner containment of the aqueous homogenous reactor experiment at ORNL in the early 1960s, a reactor that operated at much lower neutron flux than the aqueous ATW. It is not evident that LANL has addressed this problem.

Pressure-Tube Failure

Fluid-fuel boundary-layer heating is more severe the higher the neutron flux. In the aqueous ATW concept, the failure of a pressure tube can lead to pressurized hot fuel slurry ejecting into the surrounding moderator or steam generator. In all pressure-tube reactors, such as the CANDU reactors and the N reactor at Hanford, failure of a pressure tube and ejection of pressurized hot coolant into the surrounding moderator has been a foremost consideration in accident analysis, even though those reactors are not subject to fissioning boundary-layer heating at the pressure-tube surfaces. Thermal shocks and failure of nearby pressure tubes are major concerns, even for a nonradioactive coolant. This is a more acute problem for the aqueous ATW because of the fissioning boundary-layer heating and the highly radioactive, hot pressurized fluid fuel. It increases the possibility of significant reactivity increases if fluid fuel is ejected into the moderator region. The whole of the heat exchange systems are subject to the troubling effects of slurry and solution operations in the heavy-water process fluid in the four loops.

Explosive Gases From Radiolytic Decomposition

As explained above, large quantities of a stoichiometric mixture of deuterium and oxygen gases are expected to be generated from radiolytic decomposition of the heavy-water carrier of the fuel slurry in the aqueous ATW. The radiolytic gases will be highly contaminated with radioactive products from fission and neutron activation. Gas pocketing within the reactor core could result in reactivity transients, and the gases must be rapidly removed and recombined to avoid chemical explosions. This problem led to the design of in-core centrifugal gas–liquid separation and on-line recombination of the extracted gases in the aqueous homogenous reactor experiment at ORNL in the early 1960s. Note that high concentrations of radiolytic O_2 and H_2 can potentially accelerate oxidation and hydrogen pick-up of hydriding problems in Zircalo components.

Comparative Thermal-To-Electrical Efficiencies

The parameters for the various ATW concepts are displayed in Table F-3, including their thermal power, the electrical power required by the accelerator, and the net electrical power for delivery to the electrical grid. For the ATW-1 (aqueous) concept, the rated thermal power is 8,300 MWt, and the rated net electrical power is 1,601 MWe. Thus, the net thermal-to-electrical efficiency for the ATW-1 concept is 19.3%. This compares with 33-36% for the net thermal-to-electrical efficiency of the nonaqueous ATW concepts, which is comparable to those of the LWR and ALMR reactor concepts.

The accelerator to power the ATW-1 consumes 889 MWe at rated capacity producing a beam of 250 mA at 1,600 MeV, as shown in Table F-5, with a wall-plug efficiency of 0.45 assumed by LANL. Thus, the gross electrical power at rated capacity is 2,490 MWe for a gross thermal-to-electrical efficiency of 30%, the accelerator in the ATW-1 consumes about one-third of the gross electrical power. For the nonaqueous ATW concepts, the accelerator consumes a considerably lower fraction of the electrical power. For example, Table F-3 shows that the nonaqueous LWR-fueled ATW-2 produces a net electrical power of 2,180 MWe at rated capacity, which is 36% higher than that of the ATW-1, while the smaller ATW-2 accelerator requires only 44% of the beam current of the ATW-1 accelerator at half the beam voltage.

The gross thermal-to-electrical efficiency of the aqueous ATW is a consequence of the calandria pressure-tube design, which can be compared with the design parameters of the CANDU heavy-water-moderated commercial reactor, which originated the calandria design. To improve the efficiency of thermal conversion, LANL has designed for much higher pressure and temperature of the ATW fuel by thickening the walls of the Zircaloy pressure tube. However, thickening the tube walls reduces the fraction of neutrons that reach the TRU slurry within the pressure tubes from the heavy-water bath in which the neutrons are moderated. This results in lower neutron multiplication and fewer neutrons available for transmutation, for which LANL compensates in part by increasing the slurry density. Thus, the design for more efficient thermal conversion is far less neutron-efficient than the nonaqueous ATW design, which operates at high temperature with no neutron-absorbing structure between the graphite moderator and the fissioning fuel.

Operations Of ATW-1 As a Just-Critical Reactor Without Accelerator

If the ATW-1 concept were to operate as a just-critical reactor without an accelerator, the net thermal-to-electrical efficiency would be 30%, about 50% higher than for the accelerator-driven concept proposal by LANL. Presumably, however, the accelerator-driven ATW-1 would not be able to charge more for electrical energy than any other large-base load source. The lower return on investment underscores the cost penalty for operating an accelerator. To illustrate the difference in total system cost, the committee considers a campaign to process and transmute all the 60,000 Mg of TRUs contained in the LWR spent fuel accumulated by the year 2020, which are otherwise destined for a geologic waste repository. For a rough estimate the committee assumes that the unit costs are constant, are independent of the blanket thermal power, and are roughly the same for the various ATW concepts, including a just-critical modification of the ATW-1 concept.

Using the "at capacity" data of Table F-3, the actinide burn rate of 2,602 kg/yr requires 23,060 reactor years to consume the 60,000 MgHM originally in the spent fuel plus the additional TRUs produced. If the TRUs are transmuted by a system of accelerator-driven ATW-1 reactors with a net electrical power per reactor of 1,281 MWe at 80% capacity factor, the total electrical energy available for sale would be about 2.6 × 10^{14} kW·h. If the ATW-1 were operated as a just-critical transmuter without the accelerator under the assumptions of the simplified estimate, the net electrical power per reactor would be 1,992 MWe at an 80% capacity factor, so that the total electrical energy available for sale would be about 4.0 × 10^{14} kW·h. Assuming that electrical energy could be sold to the grid for about 2 cents per kW·h, the system cost differential in changing from an accelerator-driven to just-critical ATW-1 would be about $2,800 billion. Additional savings would accrue from not having to construct and operate the accelerators.

Clearly, the ATW-1 pays heavily for using an accelerator if the sale of electrical energy is expected to contribute significantly to funding the construction and operation. To realize the potential advantages of using aqueous reprocessing technology, which is more highly developed than that used in the nonaqueous ATW, the features of a just-critical aqueous ATW without accelerator should be carefully examined by LANL. Of course, the very high fissile specific power associated with the thermal neutron flux, 3 × 10^{15} neutrons/cm^2·s—and the attendant high rates of radionuclide destruction and heat production—would present materials and engineering design challenges for fuel and reactor proper, with or without the accelerator. Nonetheless, LANL may find that eliminating the accelerator from the nonaqueous ATW could result in a simpler and a more economical design.

State of the Technology

This section evaluates the state of the technology and outlines what needs to be done to further develop the various subsystems of the ATW approach. As in previous sections, we start with the accelerator and proceed through the several components and assemblies.

Accelerator System

The accelerator requirements of the ATW concept proposed by LANL are attainable with extrapolations of existing technologies. The ATW linac is made possible by the existing technology base at Los Alamos Meson Physics Facility (LAMPF), technological advances fostered by the Strategic Defense Initiative, Neutral Particle Beam program, and the improvement of high-power RF sources. A 1989 Energy Research Advisory Board stated that "the continuous-wave RF linac approach for APT is technically sound." A subsequent JASON review determined that "the technology base exists to allow the accelerator to be built with high confidence."

Critical components of the ATW system such as ion sources, RFQ accelerating structures, and DTLs have been demonstrated at a number of facilities such as LANL, Chalk River, and ANL. A demonstration of a single-leg funnel was carried out at LANL. An integrated demonstration of a high-current, low-energy (less than 40 MeV) facility was recommended as a critical component of the development process, and in 1992 DOE funding for a conceptual design of a high-power accelerator for tritium production was initiated. Many of the results from this effort would be directly applicable to the further design and development of the ATW system. In addition, the operating experience of the LAMPF for about 10 years at 1 mA (0.8 MW), provides a solid basis for potential ATW test and demonstration facilities.

This is especially true in the area of beam losses. In order to assure hands-on maintenance at 100 to 200 mA, it is necessary to limit losses to 1×10^{-8}/m, a demanding performance requirement. However, actual operating experience at LAMPF and similarity of the ATW beam physics to LAMPF operating regime provides a good basis for controlling beam losses. In addition, design features of the ATW linac can assure lower loss fractions than in LAMPF and increase the likelihood that these fractional beam losses can be achieved. Nevertheless, the design should include some provisions for remote handling, especially in the area of the emittance filter, beam line, and target.

The existing technology base applicable to the ATW accelerator system's specifications and requirements has the several components illustrated in Figure F-7.

- *Injector and RFQ*. A proton source is operating at Chalk River that has produced a proton output of 105 mA, with an RMS emittance of 1.2×10^{-6} m-rad. With modest extrapolation, this would meet ATW performance requirements. In addition, an output of 70 mA continuous wave has been achieved at Chalk River with a 600-keV RFQ (267 MHz) injected by an ECR source.
- *Drift Tube Linac (DTL)*. CERN and other high energy physics laboratories have operated a 100-MeV, 201-MHz proton DTL that reaches pulsed currents as high as 250 mA as an injector. At Los Alamos, a 7-MeV, 425-MHz ramped gradient H-DTL (pulsed, low duty factor) has achieved 100 mA as part of the Strategic Defense Initiative Neutral Particle Beam program.
- *Funnel*. A single leg funnel at 5 MeV, 60 mA, and 425 MHz has been demonstrated at LANL. This experiment confirmed 100% beam transmission with no measured increase in transverse emittance.
- *Coupled Cavity Linac (CCL)*. An 800-MeV side-coupled 805-MHz CCL currently operates at 12% duty factor at LANL. This CCL has demonstrated 17-mA peak current and an average gradient of 1 MV/m.
- *Superconducting Cavity Linacs*. Superconducting cavity linacs inherently have higher gradients and thus potentially lower capital costs. Based on the estimated RF-to-beam efficiency of 84%, a superconducting cavity linac appears to present no significant advantage in operating costs.
- *RF Generators*. At present there are 1-MW continuous wave klystrons available from industry at 350 MHz, 500 MHz, and 1,000 MHz. In addition there are new high-efficiency tubes being developed by the Institute of Nuclear Physics at Novosibirsk.
- *Beam Transport*. The beam transport system requirements for ATW are based mostly on experience from high energy physics facilities. Appropriate higher-order beam optics design, including energy spread and magnet engineering, are well understood.
- *Beam Splitter*. Several high energy physics laboratories currently utilize RF particle separators with deflection requirements that are similar to those in the ATW accelerator. However, the possibility exists that a beam-sharing scheme that involves switching on a macro

pulse basis using kicker magnets could be used. This scheme is operating at the Los Alamos Meson Physics Facility as well as other laboratories.

- *Current and Energy Stability*. Energy stability expected on target ($\delta E/E$) is $\pm 1 \times 10^{-3}$ based on Los Alamos-Meson experience and the large energy spread acceptance of the beam transport channel. Based on source performances, the expected current stability ($\delta I/I$) is $\pm 1 \times 10^{-2}$ for a continuous wave accelerator.

Physics Issues

Assuming a reference system design based on room-temperature accelerating structures, there appear to be no beam physics questions or concerns that would prevent the achievement of the desired parameters (250 mA continuous wave at 1,600 MeV). Peak currents and RMS emittance values are within the range already attained in low-duty-factor injector linacs that are operating at high-energy physics facilities. They are also consistent with values demonstrated within the Strategic Defense Initiative's supported Neutral Particle Beam program. There are currently klystrons to provide high-power continuous wave RF (0.5 to 1 MW) within the required frequency range. In comparing the existing LAMPF CCL to the ATW accelerator, LANL presented the following values:

	LAMPF	ATW
Protons per bunch	0.52×10^9	2.23×10^9
Current	1 mA (average)	250 mA (average)
Duty factor	6%	100%

These figures are significant because the protons per bunch is one of the most critical factors in determining the state of technology. Extrapolation from these values indicates that while the average current in the ATW linac is 200 times greater than that of the highest-power existing linac (LAMPF), the charge per bunch is only 4.3 greater.

The primary concerns with the ATW accelerator relate to system performance at high-power, long-term continuous operation and high efficiency in power-grid to beam-power conversion. There are currently no accelerator systems operating under continuous wave conditions at or near the very high beam power level specified for the ATW machine. There are some other technical issues that have been identified as requiring additional attention. They are

1. *Funneling*. This is a key element of the ATW linac reference design, since it reduces the injector and RFQ current specification by a factor of 2, which is leaving the charge/bunch in the CCL the same. There is no long-term operational experience with funneling, but a funneling experiment has been done at LANL, and the results were as predicted by simulation.

There are still some concerns about funneling, which include the effect on the bunch longitudinal tails of the nonconstant fields in the RF deflector. Sensitivity to momentum variations in the incoming beams should be slight because of the large momentum acceptance of the funnel as compared with the beamlets.

2. *Beam Loss in the Accelerator*. Beam losses must be kept to $1 \cdot 10^{-8}$ /m operating at 100 to 200 mA in order to allow hands-on maintenance of the machine. However, design of the ATW linac should include compatibility for remote handling throughout the system, in case it is needed, and in the region near the emittance filters and targets, where activation levels will be much higher. In this region, remote handling is almost certain to be necessary.
3. *Operational RF Control*. This is an area for major design concentration due to high beam-loading and high power levels. Feedback and feed forward systems must be designed to assure a precisely balanced system.
4. *Off-Normal Conditions*. Possible fault modes and transient behavior must be carefully studied and appropriate response systems developed in order to protect the accelerator and its power supply. The beam can be turned off in response to a number of off-normal conditions within 20 MA using a fast-abort system. The accelerator will be designed incorporating a variety of detection and protection devices in order to defend against off-normal conditions.
5. *Reliability of Components*. The reliability of components must be such that the goal of 85% availability can be achieved. This can be helped by equipment redundancy, rapid access to critical spares, regular maintenance, and identification of susceptible components prior to failure. It is also valuable to have electronic components in a shielded area to allow access to them while the accelerator is operating.

Aqueous Atw-1

Feed material for the aqueous ATW could be prepared by conventional PUREX aqueous reprocessing, supplemented by TRUEX, or another three-valent element processing system, to achieve high-yield recovery of TRUs and fission products. As noted elsewhere, the TRUEX process is not yet sufficiently developed for commercial or production application. Additional laboratory-scale, pilot-scale, and prototype operations would be needed. To obtain the economy of large-scale aqueous reprocessing considered necessary for U.S. construction, an annual reprocessing plant throughput of about 600 Mg/yr of LWR spent fuel would be required, which is near the size of the large reprocessing plants currently under construction and start-up worldwide. This could furnish the TRU feed for about 15,000 MWt of ATW power for the ATW concepts that transmute only TRUs and fission products from LWR spent fuel. This would require a commitment to build a significant number of ATWs. Thus the aqueous ATW could rely on existing and near-term reprocessing technology to obtain its start-up and make-up TRUs from LWR spent fuel.

However, the on-line reprocessing proposed for the aqueous ATW would require the development of many new separation technologies. Intense radiation levels are expected because of the short cooling times, the multiple recycle of TRUs and fission products, the many different separations required for the TRUs, the fission products, and the accelerator target.

Also, economical technology on the small scale (100 to 200 tons of HM per year of a single integrated on-line processing system for each ATW) will be difficult to achieve, especially with the many special steps required. Separations technology developed for the aqueous homogenous reactor is of questionable value. The ATW fluid fuel of TRU oxide particles suspended in heavy water differs chemically and physically from the solution of uranyl sulfate in heavy water in the aqueous homogenous core and the thorium-oxide suspension in the blanket of the ORNL experimental reactor.

In any calandria reactor, the escape of high-pressure hot coolant into the relatively cool low-pressure moderator outside the pressure tubes, through a possible rupture of a pressure tube, is a serious safety issue. It would be far more serious in the fluid-fuel aqueous ATW because of the extreme radioactivity of the fluid fuel and because large increases in nuclear reactivity could result. As an additional precaution, LANL proposes that each calandria tube be two concentric Zircaloy pressure tubes, each capable of withstanding the pressure difference between the hot fluid fuel and the relatively cool heavy-water moderator.

Pressure tubes in the CANDU calandria structure are also of Zircaloy. Because neutrons thermalized in the low-pressure heavy water outside the tubes must diffuse through the pressure-tube walls to cause fission, it is necessary to limit the pressure and temperature within the pressure tubes and to maintain small-tube metal inventories to avoid excessive absorption of neutrons by Zircaloy. Neutron absorption in the pressure tubes would be a more severe problem in the aqueous ATW design than in the CANDU, because neutrons must diffuse through the walls of two concentric Zircaloy pressure tubes. Consequently, LANL designs for much smaller pressure tubes (about 5 cm in diameter) than in the CANDU. This requires a much larger number of pressure tubes to obtain the same thermal power. The calandria in the ATW is thus a far more intricate structure than that in the CANDU.

The CANDU reactor itself operates at higher neutron flux than commercial LWRs. Radiation damage to the Zircaloy pressure tubes requires tube replacement after about 15 years of operation, a demanding and difficult maintenance operation. Because of the tenfold higher neutron flux of the ATW, radiation damage is accelerated, requiring tube replacement every 1 to 2 years. Hence, decontamination and disposal of radioactive structures may be an important waste disposal issue.

The fluid fuel of the aqueous ATW is a suspension of oxide particles in heavy water along with fission products like CsI in solution or colloidal suspensions. Data on similar slurries indicate that this slurry will erode the steel and Zircaloy of pressure tubes and piping more severely than does the chemically optimized, heavy-water coolant of the CANDU reactor. Erosion can be reduced if small particle sizes are maintained. LANL contemplates fluid-fuel slurries with particulates of about 10 microns in diameter. These are smaller particles than were used in the slurry blanket of the aqueous homogenous reactor experiment at ORNL in the early 1960s, and they will become smaller on the average as fission proceeds.

Reactivity fluctuations due to variations in slurry density have always been a concern for reactor concepts fueled with slurry fuels. Other than a low-power experiment in Holland decades ago, there has been little experience with slurry fuels. With such small fuel particles, many of the fission fragments can be expected to recoil into the heavy-water carrier, which results in radiolytic decomposition of water and production of a stoichiometric mixture of deuterium and oxygen gas, as well as changes in water chemistry and viscosity by fission products as mentioned earlier. A typical ATW blanket generates 2,080 MWt at full power. For an estimated 50% escape of fission fragments from fuel particles, and assuming 200 MeV of fission-fragment energy to decompose a heavy-water molecule, about 1.5 m^3/s (at standard temperature and pressure) of decomposition gases would be produced. The gas would be entrained in the fuel slurry that flows turbulently through the pressure tube. The gas would have to be rapidly separated and recombined to avoid explosions and detonations and to avoid gas pockets being entrained back into the reactor core. Although deuterium and oxygen radiolytic gas was formed and recombined in the aqueous homogenous reactor experiment, and although a relatively small amount of hydrogen and oxygen result from gamma radiolysis in LWRs, radiolytic gas of the magnitude predicted here has not been encountered in any operating reactor. Separating and recombining the deuterium and oxygen is expected to be a formidable problem. Techniques used in the aqueous homogenous reactor experiment, involving vortex flow in the reactor core to rapidly separate the radiolytic gases, are probably not applicable to flow in a long small-diameter process tube.

No large power reactor has operated at the neutron fluxes contemplated for the aqueous ATW. Experience has been limited to the small high-flux research reactors. Those reactors have experienced the need for careful operation to avoid permanent shutdown due to transients in the amount of ^{149}Sm, as discussed in Chapter 4 under *Safety Issues*. None of these high-flux research reactors has been large enough to experience the problems of xenon oscillation that are exacerbated by high thermal flux.

Circulating a fluid fuel to an external heat exchanger introduces new problems of fission boundary-layer heating at the inner surfaces of pressure tubes, as discussed earlier. This problem was encountered in the aqueous homogenous reactor experiment at ORNL in the early 1960s and contributed to the failure of the core container. Fission boundary-layer heating would be aggravated by the high neutron fluxes of the ATW. Current technology of nuclear power reactors has not addressed this problem. Temperature differences will cause the formation of deposits from the always-saturated solution of the slurry. Even LWRs can be very sensitive to water chemistry and the formation of deposits. On the slightly acid side, there can be a loss of several percent in pump efficiency from deposits as compared with operations on the basic side.

Nonaqueous ATW

The nonaqueous ATW would introduce new materials problems because of the molten fluoride fluid fuel and the use of lithium coolant. Lithium is more difficult to contain than is the sodium coolant used in liquid-metal reactors. Lithium was studied as coolant in one of the reactors considered for nuclear aircraft propulsion in the 1950s and early 1960s. Lithium was also adopted as the coolant in the more recent project to develop a nuclear power system for space applications. Special niobium or vanadium alloys are required for piping external to the reactor. Such alloys have not been used in operating reactors and indeed have a limited industrial experience or technology base. There is also the problem of the mass transport of metals from the hotter to the cooler parts of the system. The ^7Li required is not in the U.S.

inventory, and no U.S. plant to produce it exists; however, Russian or French capacity to produce it may exist.

Graphite in molten-salt reactors must withstand neutron irradiation, must have pores sufficiently small that capillary forces exclude fuel salt, and must have sufficiently low permeability to limit the absorption of ^{135}Xe. Although a graphite stringer from the molten-salt reactor experiment (MSRE) showed no effect of 2 1/2 years in contact with fuel salt, it would not have met the requirements for radiation damage and gas permeability of a molten-salt power reactor (McNeese and Rosenthal, 1974).

The LiF-BeF$_2$ fused salt, which contains fluorides of TRUs, is similar, but not identical to, the fused salt adopted for the MSRE at ORNL in the 1960s. There the fuel was UF$_4$, and the salt also contained ZrF$_4$. Also, higher operating temperatures and neutron fluxes are planned for the ATW.

MSRE vessels and piping were constructed of Hastelloy N, a nickel-base alloy containing 16% molybdenum, 7% chromium, and 5% iron. Reduction of creep ductility of Hastelloy N by neutron irradiation was not a problem in the MSRE, but was expected to be an issue for a power version. Development of an improved alloy for a power reactor was underway but was terminated. Hastelloy N showed generally low corrosion rates, but intergranular surface cracks were observed after MSRE operation. These were thought to be due to interaction with fission-product tellurium. Modifications were underway to develop an alloy unaffected by tellurium, but they also were not completed (McNeese and Rosenthal, 1974). Nickel-base alloys were abandoned from fast reactor and fusion program materials considerations because of ductility loss to high fluence.

Radiation damage in most graphites results in shrinkage followed by expansion at neutron fluences below those of interest for power reactors operating at normal neutron fluxes. Special grades of graphite show little shrinkage and a longer period before rapid expansion begins. Far more must be known about the radiation damage behavior of graphite at ATW temperatures in order to predict clearances needed and frequency of replacement.

In the MSRE, the fused salt was in direct contact with graphite moderator, as is planned for the ATW. Sealing graphite to exclude xenon could involve the use of pyrolytic carbon deposited in the surface pores or in a thin layer on the surface. Test samples for the MSRE showed excessive increase in permeability under neutron irradiation. New preparations were under way when the program was canceled (McNeese and Rosenthal, 1974). Xenon in graphite is a crucial issue for the nonaqueous ATW because of neutron absorption by ^{135}Xe and because of the danger of power instability resulting from xenon oscillations. There will be a small concentration of fluorine in the melt from the irradiation to cause slow corrosion of almost everything in the system.

Some development of chemical processing of LWR spent fuel by nonaqueous halide techniques has been done for special fuels in a program of the DOE, but not at the scale and recoveries needed for the ATW. Its application as an integrated, small-scale, head-end facility for one or a few ATW reactors as proposed by LANL may not be economical. The on-line centrifuge separations proposed for recycling TRUs and fission products would require very extensive and expensive development, including proof of concept. No machines of the required materials, which will probably need to be composites, have ever been developed.

Target Materials

There is limited experience with materials performance at "spallation" sources of this kind (LAMPF at LANL, SNQ at KFa Julich, and SINQ at the Paul Scherrer Institute), and the proposed conditions for sources in the ATWs are at fluxes and fluences greater than these experienced in these facilities. Moreover, the bulk of the experience to date has been with solid targets, primarily copper. Use of mixed low and high Z targets with differing thermal conductivity will require extensive testing and demonstration. The use of liquid-metal targets removes the concern over displacement damage to the target (but not to the structure containing the liquid target). Moreover, liquid-metal targets introduce concerns over corrosion and erosion of containment structures. Performance of these systems in a high-flux, fluence, high-energy neutron/proton environment remains an unresolved engineering challenge. There remain the problems of the heat exchangers for the liquid targets and the chemical changes introduced by the spallation of the target elements.

Blanket Assembly

The aqueous and nonaqueous ATW subcritical reactors, and their associated on-line reprocessing systems, represent considerable extensions beyond existing technology. The committee has not been presented with a thorough and detailed estimate of the new problems to be solved, since extensive experiments have yet to be performed. At this early stage of design and development, the committee can do little more than identify examples of the many technical issues that must be resolved. Many of the issues of new technology development can be best understood by comparing the proposed ATW technology with technology of well-developed existing nuclear power reactors.

Compared with modern solid-fuel nuclear power reactors, both the aqueous and nonaqueous ATW concepts would encounter severe material problems and new safety problems because of the very high thermal neutron fluxes, the use of fluid fuels and fluid targets, the high power densities, and the use of continuous integrated chemical processing with short turnaround times. Examples of problems associated with each of the two concepts have been suggested earlier and are expanded.

Materials Development

ATW-1. The proponents of this system claim that by approximating the calandria structure of a CANDU reactor for the blanket moderating structure (zirconium alloy pressure tubes [containing a slurry fuel instead of fuel assemblies] surrounded by D_2O moderator), the engineering experience to date of the CANDU systems can be used to advantage. It is true that the CANDU systems have by and large operated well up to now, but they have not done so without problems. Radiation-induced creep and growth have led to premature sag of pressure tubes in some systems, and delayed hydrogen cracking has led to early failures of some zirconium alloy components. In general, zirconium-based alloys are not operated beyond a few tens of dpa (displacements per atom) corresponding to fluences in typical fission reactor spectra of 2-3 × 10^{22} neutrons/cm ^{2} (E_n>1 MeV). Although specific fluence targets are not stated in the ATW descriptions, the implication is that fluences of at least this magnitude could be achieved; this will limit blanket lifetimes to a few years at the flux levels anticipated for ATW.

Moreover, being highly thermalized fluxes, transmutation of constituents with high thermal-neutron-capture cross sections can become important in other structural materials (like aluminum alloys) that have been proposed. The transmutation-induced property changes can sometimes be more limiting than the radiation damage. Finally, irradiated materials are often much more susceptible to be induced by the chemical environment degradation, like stress-corrosion cracking, because of segregation of impurities and alloying elements or because of strength increases/ductility losses or both. Structural materials proposed for ATW-1 (particularly zirconium alloys) are known to be sensitive to halogen, OH-, and hydrogen content of the environment; hence, careful attention will have to be paid to the nature of the fuel slurry as well as water chemistry in the D_2O moderator.

ATW-2, -3, -4. These systems replace D_2O with graphite as a moderator and replace the fuel slurry with a fused salt. The behavior of graphite in a thermal neutron irradiation environment is fairly well known, and graphite has been used with success in high-temperature gas-cooled reactors. Dimensional stability is generally good at intermediate temperatures where irradiation growth is minimized and gas-induced swelling is avoided. There may be a concern with additional helium gas generation by (n, α) reactions induced by high energy neutrons (in the high energy tail), but this is probably only a potential concern near the source. On the other hand, ^{14}C generation in graphite has waste disposal implications.

There has been some experience with reactor structural materials in the presence of fused-salt fuels in the molten-salt breeder reactor program. Although high nickel alloys appear to be reasonable candidates for this application, corrosion, erosion in combination with radiation-damage induced property changes still present formidable challenges. Moreover, since the molten-salt breeder reactor program, extensive experiments in support of the fast-breeder reactor have shown that high nickel alloys are susceptible to radiation embrittlement at high fluences. Beyond this class of alloys, austenitic stainless steels suffer from dimensional stability problems (void swelling) at high fluences, and other structural alloys for high temperature application (ferritic stainless steels, vanadium alloys) are far from being well developed for this application.

Chemical Processing

Most of the development issues concerning the ATW concepts are described in Chapter 2. While the chemistry of these processes is being developed and understood, the engineering database is not available and would require extensive scaling and testing. The many issues here reflect a very immature technology. The chemical processing proposed for the ATW concepts is beyond the state of industrial experience because of both the radiation fluxes involved and the number of separations that are required to maintain any of the concepts. The reliability factors required to keep the systems on line as required are beyond the present state of the equipment art for that environment. The proposed use of high-speed centrifuges for the separations required in the molten-salt systems may be impossible, and the cycle will have to use structurally less-demanding techniques, like those pyrochemical procedures for the ALMR (IFR), combined perhaps with thermal diffusion techniques that can separate the constituents of homogeneous systems, put little mechanical stress on the materials of construction, and have simple moving parts.

Cost and Schedule

At this stage, it is very difficult to determine the cost or possible schedule of the ATW concept. The proponents at LANL have continued to support the concept and in particular, the processing, without a real charter or funds to carry out a conceptual design.

Rough schedules and costs for a conceptual design and accelerator component and subsystems have been proposed by LANL and are given in Figure F-18. This proposal indicates some $46 million over 4 years. Within this envelope a demonstration of a completely integrated ATW accelerator front end in continuous wave operation could be expected. A funneling demonstration with two beams would be an essential element to remove remaining concerns about this new system. Even given this, there remains considerable uncertainty about efficiency and long-term continuous wave operation for a system of this power and complexity.

Research and development needs for the blanket and fuel chemical processing are a much greater uncertainty. Many of the blanket elements have a history in CANDU reactors and molten-salt reactors at ORNL. On the other hand, to have the engineering database to support a reactor, albeit a subcritical one, license application is very difficult to predict and in particular, to cost. Just a simple separations pilot plant can be expected to be a billion-dollar project. This will be especially true for the proposed high-temperature, high-speed, molten-salt centrifuge system.

LANL has provided Figure F-19, which indicates that within about 6 years the flowsheets with hot and cold tests could be performed. It will likely be 6 additional years before the flowsheets could be taken to pilot scale and integrated testing, which is the earliest that any meaningfully relevant licensing data could be available. This means it may be 10 to 20 years before a construction permit under NRC standard 10CFR50 is available. Consequently, the committee expects that it will be several decades before an initial actinide or fission-product burning could take place using ATW technology. This time frame extends farther into the future than either the LWR, ALMR, or Phoenix.

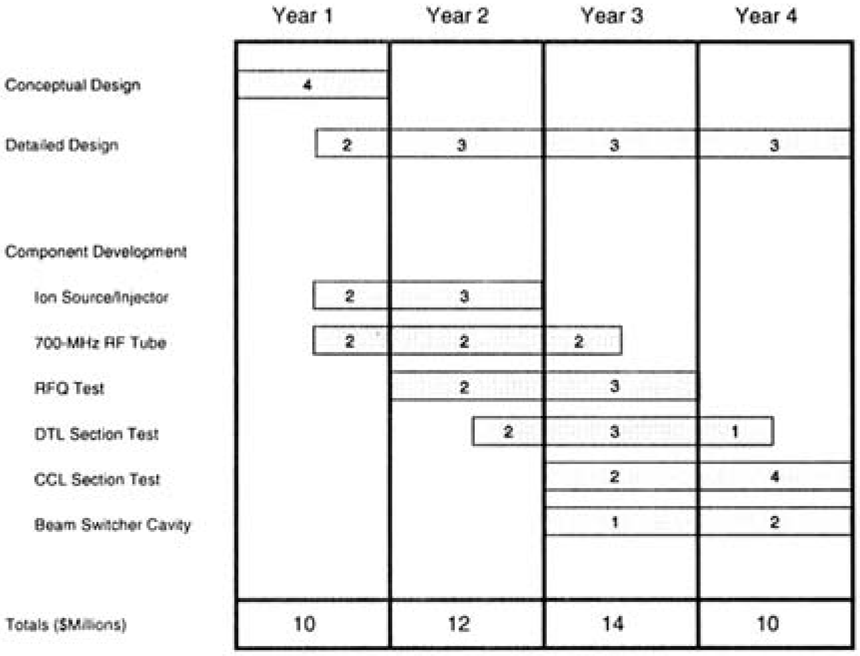


FIGURE F-18 Program for ATW accelerator design and engineering development.

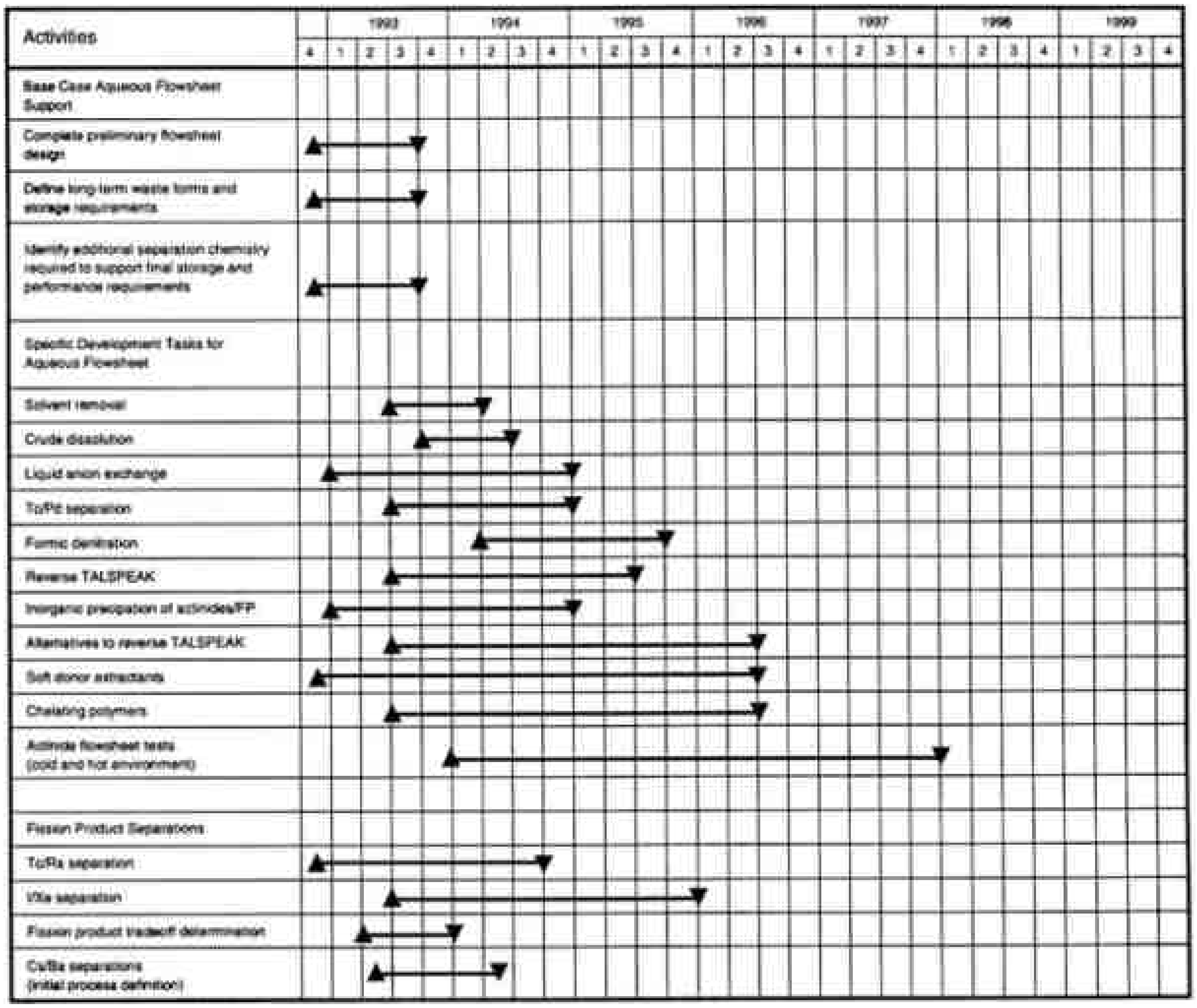


FIGURE F-19 Milestones for ATW development.

The Particle Bed Reactor Concept

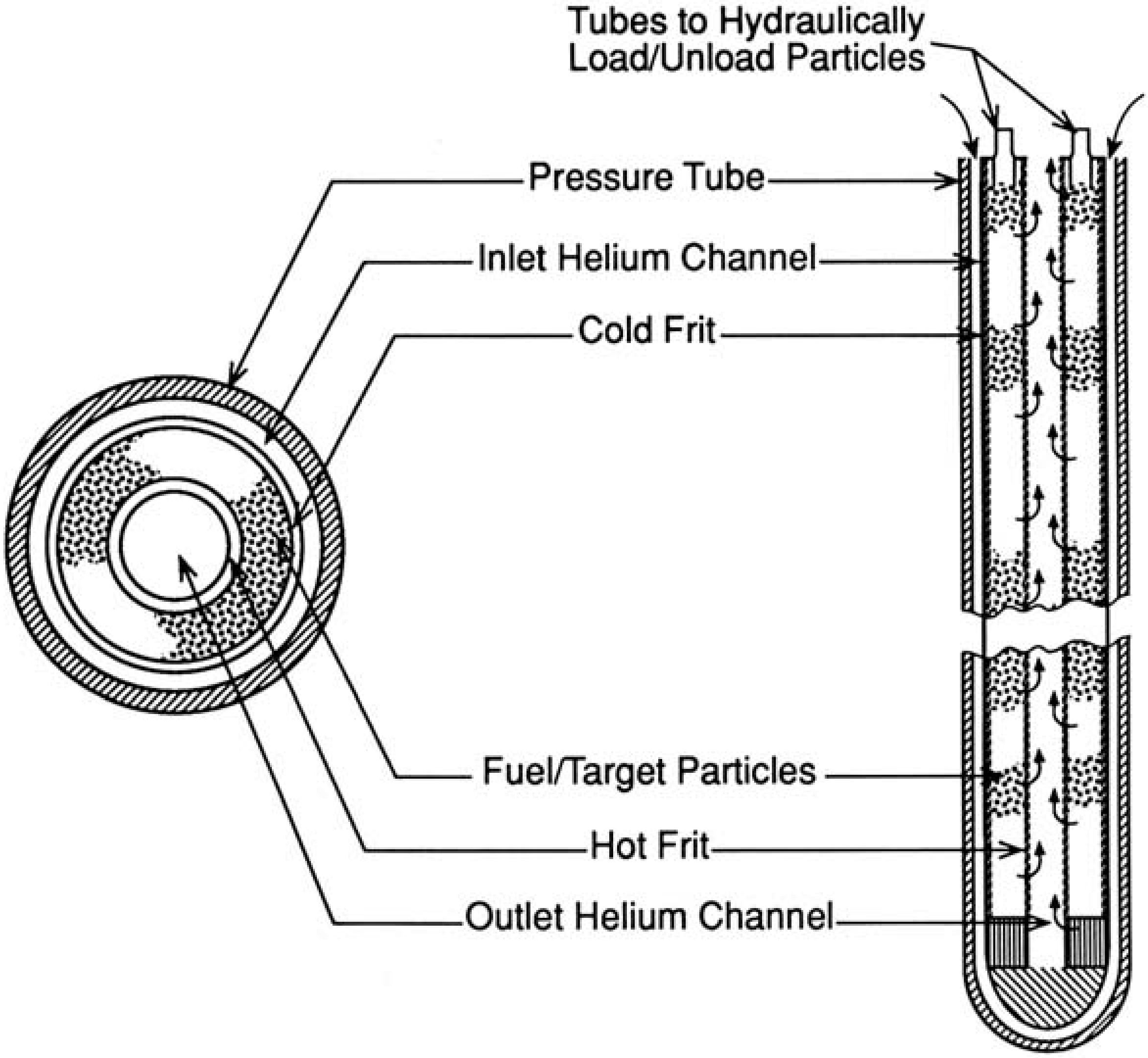
Reference Designs

Under development by Brookhaven National Laboratory (BNL) for space power applications, the particle bed reactor (PBR) was proposed as part of a system for the transmutation of HLW, with the ultimate goal of significantly reducing, if not eliminating, the long-term risk of a geologic repository. The PBR concept, which is a thermal reactor with a high neutron flux, has the potential to burn up fission products as well as the MAs. Furthermore, the reactor is meant to be compact to reduce the inventory of the actinide materials in the PBR relative to that in the LWR. The reference reactor design will be highly compact with an approximate volume of 200 liters producing a total of 1,000 MWt. The resultant power density of 5 MWt/liter is much higher than experienced in power reactors, which range from about 50 kWt/liter in boiling water reactors to 280 kWt/liter in the Super Phénix Liquid Metal Fast Breeder Reactor (Todreas and Kazimi, 1990). The power density is even higher than the 1.33 MW/liter in the high-flux isotope reactor (National Research Council, 1988). Therefore this reactor concept demands a much higher reliability of coolant flow than the current generation of reactors.

The fuel material is embedded in graphite particles that are similar to the high-temperature gas reactor (HTGR) fuel particles but have much smaller diameters (0.8 mm). Each fuel element consists of a cylindrical pressure tube in which a fuel particle bed is accommodated within two coaxial porous tubes (frits). This allows for coolant flow through the bed as well as around it. The fuel is to contain the plutonium, while the fission products and the MAs are included in similar particle beds in separate target elements. In the particles, which are porous kernels that contain 5% plutonium by volume, MA, or fission products as admixtures of graphite, are coated by impervious pyrographite.

The packed particle bed is proposed to be cooled by forced circulation of helium at 70 atmosphere pressure (see Figure F-20). The helium exits the bed at a temperature of 700°C. To remove the high density thermal energy, helium is to flow at very high velocities (275 m/s) in the central region and surrounding the particle bed, thus feeding lower-velocity helium flow within the bed. The maximum fuel-particle temperature is about 8,000°C for a design that involves a power density of 5 MW/liter. It should be noted that in the HTGR, the power density in the fuel block is only 45 kW/liter (Knief, 1981). The fuel-particle limiting temperature to prevent fission product release as proposed by BNL is about 2,200°C, which is considerably higher than the HTGR-limiting design temperature of 16,000°C (National Research Council, 1992). The calculated fuel maximum temperature of 8000°C is based on nominal fuel-particle conditions, and no allowance for agglomeration, sticking, or bridging is made. It is doubtful that such favorable uniformity can be sustained under nominal manufacturing conditions. Core thermal reliability will require quality assurance and monitoring practices that can be prohibitively costly.

As moderator, the conceptual studies involved two materials: heavy water and beryllium carbide. For Be_2C, the hexagonal moderator blocks are 1.5 m long and 0.15 m to 0.2 m wide. They are packed closely around the fuel elements, thus forming a larger hexagonal core that is



SOURCE: Brookhaven National Laboratory

FIGURE F-20 Detail of fuel/target element for waste burner.

surrounded by a reflector. For heavy water, hexagonal cans would surround the PBR pressure tubes. The moderator as well as the structural materials within the core are subjected to very high fluences, on the order of 10^{23} neutrons/cm^2. No data are available on the consequences of fluences of such a high magnitude. Gaseous products, either tritium or helium, will be produced in the moderator and need to be accommodated by specific designs that have yet to be considered.

The characteristics of burn-up (about 30 to 50%) in the small plutonium volume (5%) compare favorably with the smaller burn-up (5%) in the larger volume fraction (25%) in the HTGR particle. Because the ultimate fission product pressure in the fuel particle will be much smaller than that of the HTGR particle, a higher reliability is expected by BNL for the integrity of the particle during operation. However, because the particle coating is thinner than that of the HTGR, the stress within the coating may not be smaller. No assessment of the state of the stress in the pyrographite was presented. Therefore, the fuel-particle integrity under expected burn-up conditions is yet to be determined.

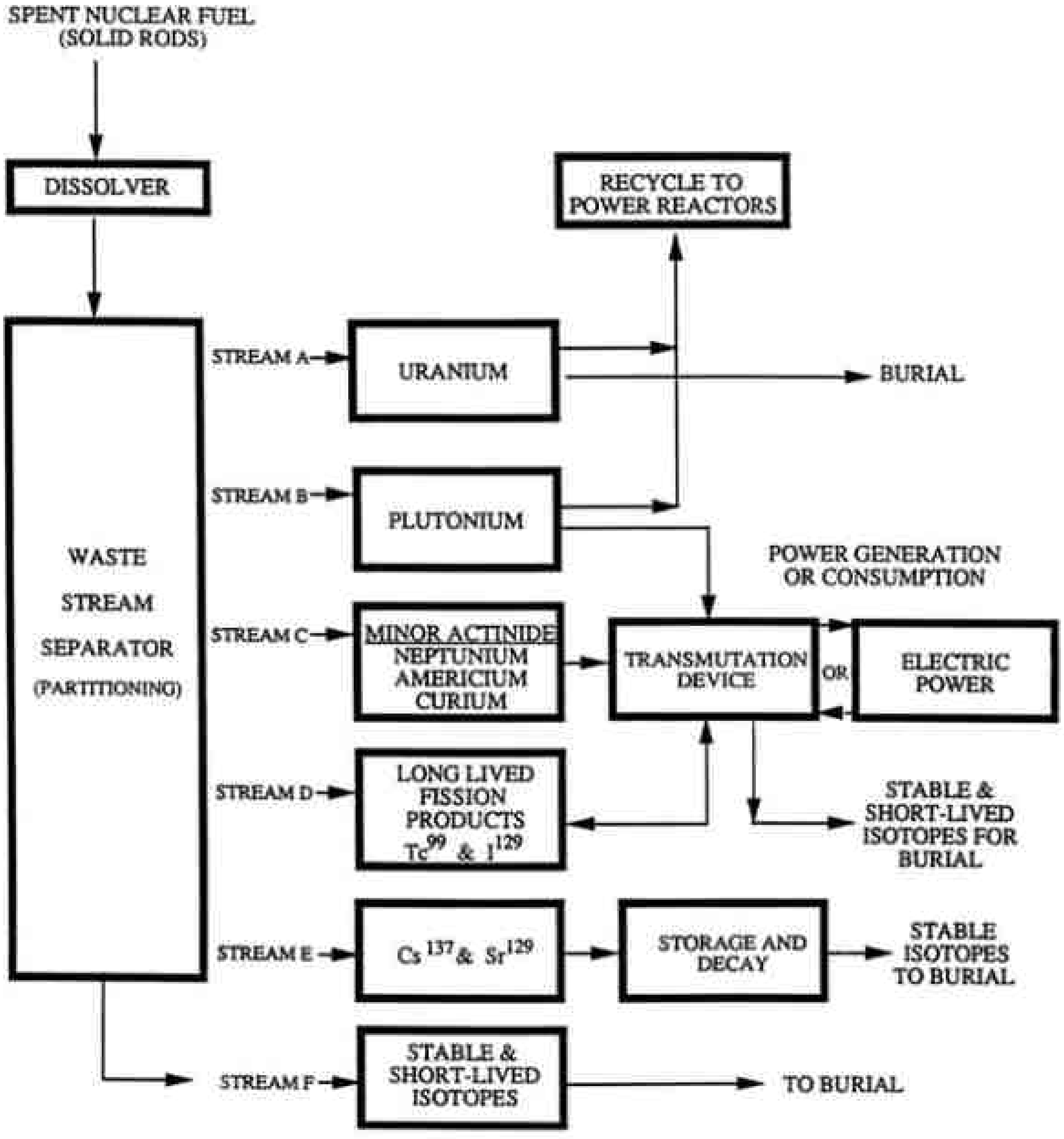
Transmutation Performance Requirements

The PBR was proposed by BNL as a transmutor for the MAs, some of the plutonium, and long-lived fission products from LWR spent fuel. The remaining plutonium is meant to be used in future reactors that accept MOX fuel. The transmutor will thus fit in a total system that aims at reducing the waste management requirements to containment of the relatively short-lived fission products, either in monitored, retrievable storage facilities or in LLW disposal sites. The system is shown in Figure F-21. The objectives of the system and a general description of its components were presented at two meetings with the Subcommittee on Transmutation. No written reports or other publications about the objectives of this concept exist. While the PBR is not meant to be necessarily a power producer, BNL staff have not ruled out the possibility of having the PBR as a net electric power producer.

The degree to which plutonium may be burned in the PBR is a variable that depends on the designer. BNL calculations for the reference design have shown that about 25% of the plutonium emerging from the power reactors may be consumed in the transmutor. This assumes that in the future other reactors will be able to use the unburned plutonium as fuel. In principle, the PBR can be designed to burn all the plutonium, but such a case was not considered by BNL.

To achieve the desired goal of eliminating the need for a repository, BNL proposes an overall separation efficiency of 10,000 to 100,000 for the removal of plutonium, MAs, and long-lived fission products. These high separation efficiencies have yet to be demonstrated for any of the separation processes proposed on a production scale.

A basic feature of the proposed PBR is a low inventory of plutonium and MA and a very high burn-up rate. Within 20 days of operation, its plutonium content decreases from 30 to 9 kg, while its MA content decreases from 18 to 11 kg. This raises a question about the reactivity swing in this cycle and the possible effects on safety and control. BNL estimated a reactivity decrease of 15% $\Delta k/k$ over a period of 14 days of burn-up. Therefore, the PBR will likely require significant control devices. If extensive burnable poison is to be provided initially in the



Source: Brookhaven National Laboratory

FIGURE F-21 Simplified overall flowsheet for nuclear waste burning.

core to offset the prevalent positive reactivity in the fuel, the tendency of the temperature coefficient in the core to become positive may be of concern, as it could lead to escalating power when the temperature of the fuel rises due to a power-to-flow mismatch.

The PBR is assumed to shutdown for 8 days to allow for refueling. This is based on the design of a very rapid particle release mechanism to achieve both defueling and refueling. However it may not be possible to process the spent fuel until it has cooled off. This means that the out-of-core inventory of the PBR fuel will be larger than the in-core inventory. A very fast process for particle recharge will require adequate controls to prevent any criticality events. This was not discussed in the design presentation to the STATS Subcommittee on Transmutation. The burn-up and recharge periods discussed above will enable the core to have 15 cycles per year.

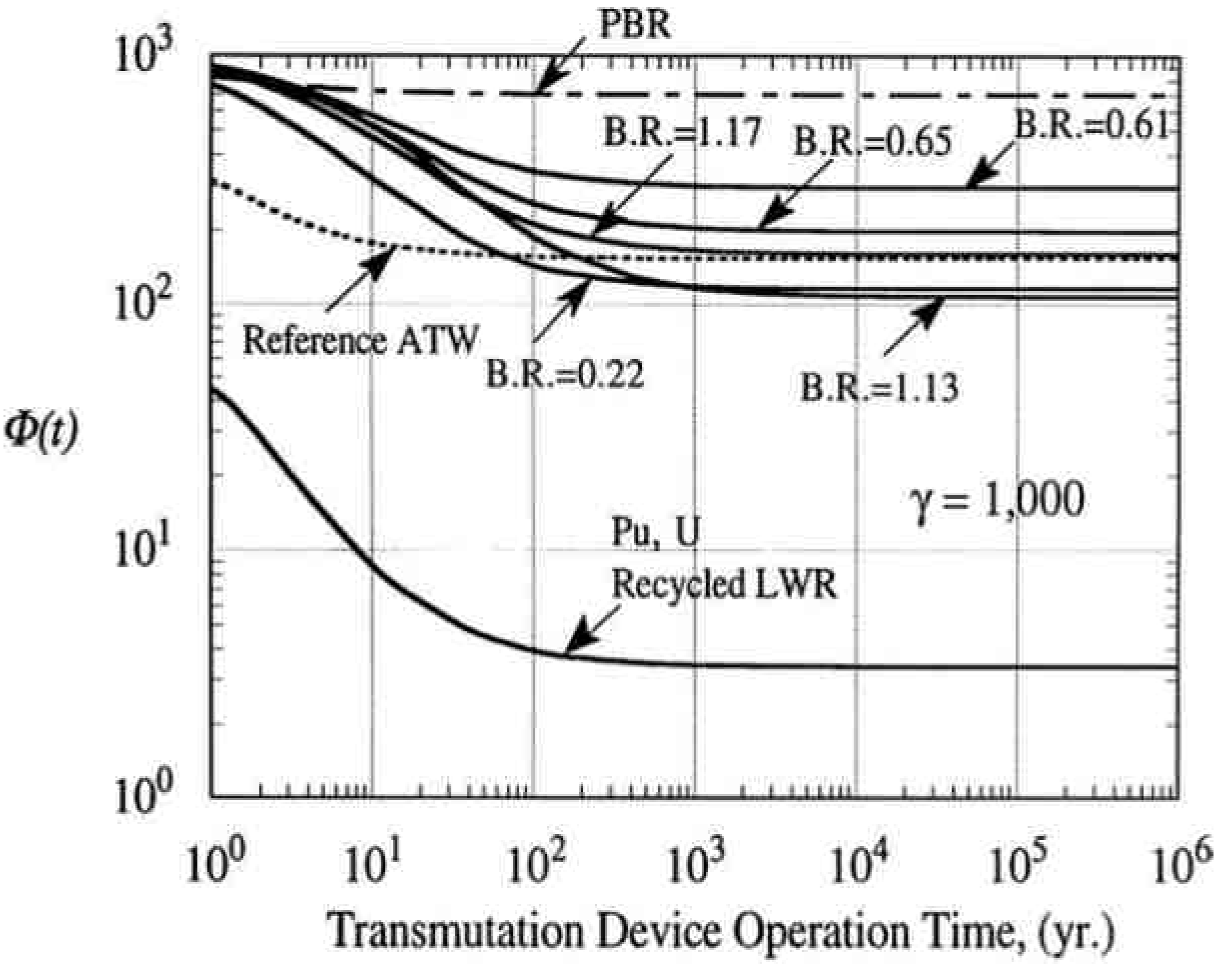
The total system inventory, meaning the in-core and out-of-core actinide inventory, is said to remain comparable with that in the LWRs it supports, since the system does not add significantly to the LWR inventories. Based on the consumption of total MA, the PBR can support five LWRs. BNL estimates refueling to take only 8 days, allowing for 15 cycles per year. If reprocessing is to be done on actinide fuel that has been out of the core for 1 year, the out-of-core inventory of plutonium will be 135 kg and MA will be 165 kg. This total inventory of about 300 kg is equivalent to the annual rate of TRU discharge from a single LWR.

Thus, if the PBR functions as proposed, it will be capable of reducing the inventory of actinides at a rate faster than the integral fast reactor, but comparable to the ATW. If the assumption is made that the PBR should accommodate all the discharged plutonium from the LWRs and be used for electricity generation, it is possible to define the inventory reduction factors for all three concepts both in the total system (see Figure F-22) as well as in the repository (see Figure F-23) (Kang and Kazimi, 1993).

Facilities and Support Requirements

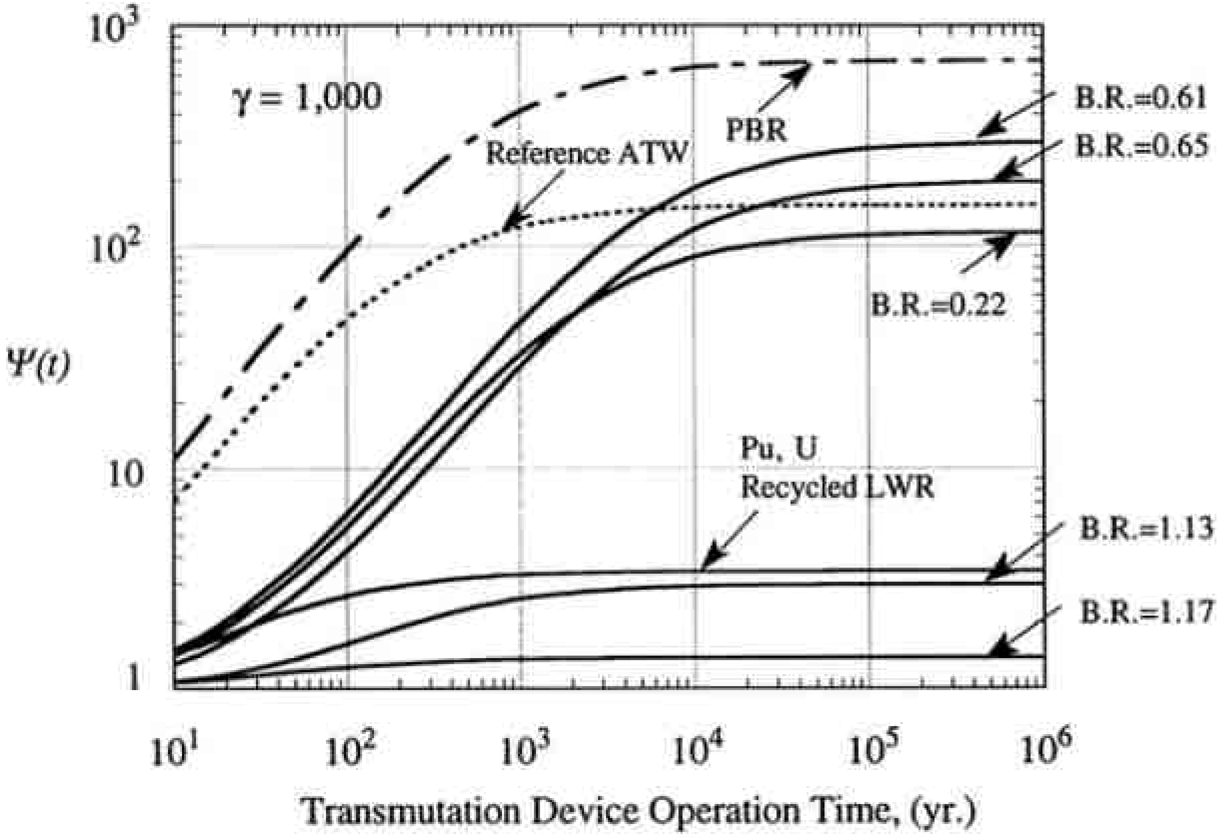
The PBR is designed to fit in a total nuclear energy system that would involve facilities with the following capabilities:

- LWR reactors that use both uranium and plutonium as fuel;
- PBR transmutors;
- MOX fuel fabrication facilities (for the LWRs);
- PBR fuel and target fabrication facilities;
- chemical reprocessing facilities that are capable of producing separate streams from the reactor spent fuel for (1) plutonium, (2) minor actinides, (3) technitium and iodine, and (4) cesium and strontium;
- chemical processing facilities for the PBR fuel-producing streams that are similar to those streams of the LWR fuel;
- monitored retrievable storage facilities for the short-lived fission products; and
- LLW disposal facilities.



SOURCE: Kang and Kazimi (1993)

FIGURE F-22 Transmutation device operation time (total system).



SOURCE: Kang and Kazimi (1993)

FIGURE F-23 Transmutation device operation time (repository).

BNL proposes that separation of the identified streams from the LWR spent fuel be accomplished by using the PUREX and TRUEX aqueous processes. The plutonium and uranium streams are then sent to LWR fuel fabrication facilities, except for part of the plutonium that is needed to fuel the PBR. The MAs and fission products are sent to the target processing facility. The PBR fuel is first sent to an electrodissolver to remove the carbon, which is then sent to the fuel fabrication facility. The decarbonized fuel and target materials are then sent to the PUREX and TRUEX processes and to a lanthanum separation facility. These steps are illustrated in Figures F-24 and F-25. The lanthanum separation process is a difficult problem with significant containment and handling requirements.

Alternatively, a modified fluoride volatility process is suggested for both the oxide and carbide fuels, in which volatile fluorides can be separated by distillation, and nonvolatile fluorides are separated through molten-salt electrolysis, laser photochemistry, or aqueous separation. This process is illustrated in Figure F-26. Again, such fluoride processes have been plagued with difficulties in the past.

Residual Waste Management and Environmental Impacts

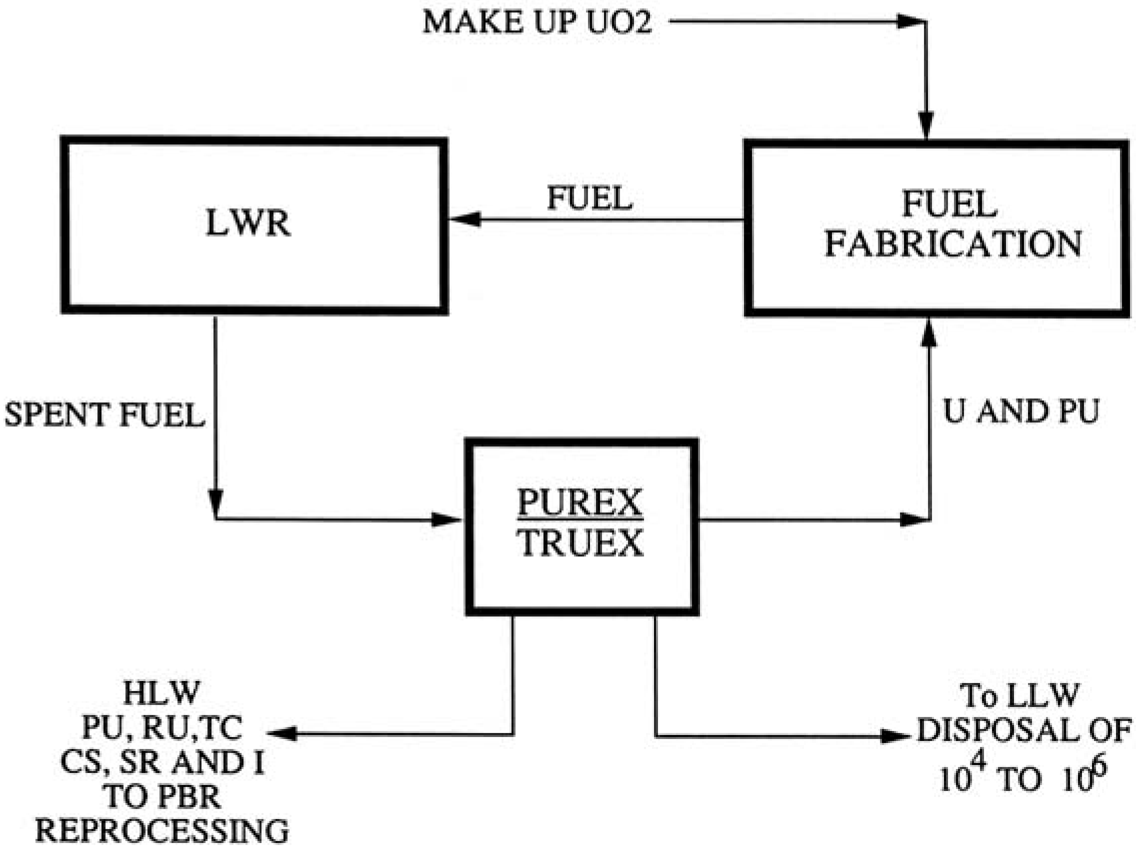
The wastes generated by the PBR will differ significantly from those in the LWR because of the differences in the fuel and in the structural and coolant materials. A large volume of graphite will be involved as well as the beryllium or D_2O moderator. The resulting wastes have yet to be assessed. Also, the fate of curium, which was found to buildup in inventory during this short burn-up cycle by a factor of 10 to 20, has not been investigated. One of the untreated products is ^{135}Cs, which will accumulate with other fission products during the burn-up cycle (a total of 13.8 g/20-day burn-up cycle at a flux of 1×10^{16} neutrons/cm^2·s). Finally, production of ^{14}C from nitrogen impurities in carbon-bearing materials would also be of concern.

Safety and Risk Considerations

At this stage of the conceptual development, there is little information about the safety features of the PBR, its dominant risk factors, or its environmental impact. The BNL staff believe that relative to an LWR, the total radioactivity in the PBR will be small (less than 10%). In addition, while this is a high-temperature reactor, its fuel is capable of handling much higher temperatures than the LWR fuel.

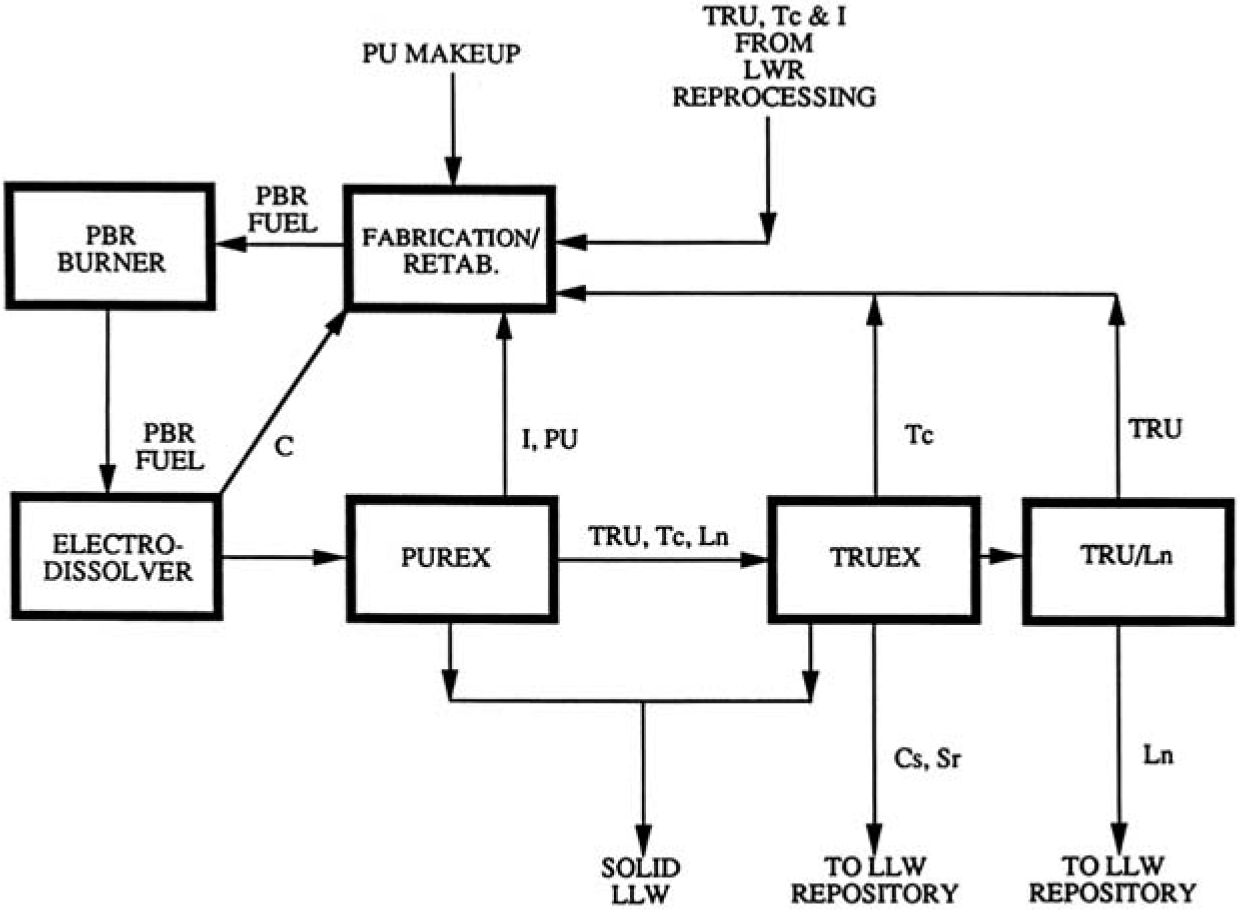
The two safety-specific features proposed at this stage are an auxiliary heat-removal loop that is in continuous operation in parallel with the main coolant circuit and a heat-pipe-cooled core-catcher below the vessel to catch any particles or fuel material that may fall from the core in overheating accidents.

It is not clear how the core would respond to any event that may interrupt the flow of the helium coolant. Such events may be due to loss of electricity supply or due to loss of coolant-pipe integrity. At high power density, it may be more difficult to ensure that adequate means of core cooling can be provided in time to prevent excessive heating of core materials.



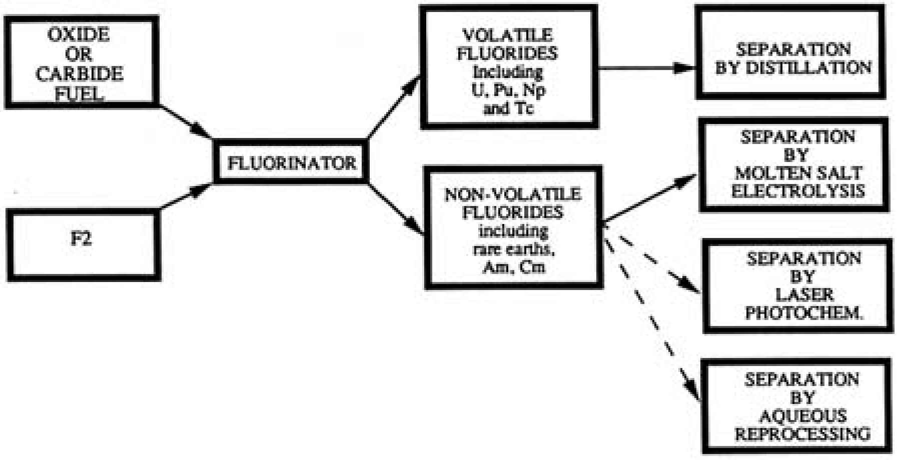
SOURCE: Brookhaven National Laboratory

FIGURE F-24 PUREX/TRUEX reprocessing for LWR fuel.



SOURCE: Brookhaven National Laboratory

FIGURE F-25 Reprocessing flowsheet for PBR and LLFP burner.



ADVANTAGES:
1. NON-AQUEOUS - NO LARGE VOLUMES OF CONTAMINATED LIQUIDS
2. EASY SEPARATION OF U AND PU
3. Tc PARTITIONING STRAIGHT FORWARD
4. NO SOLVENT DEGRADATION PROBLEMS

SOURCE: Brookhaven National Laboratory

FIGURE F-26 Conceptual flowsheet for modified fluoride volatility process.

Should the PBR not produce electricity, it may be possible to site it in remote areas and therefore not pose a significant risk to populated areas. (This will be at the cost of losing the revenues that may be realized from sale of electricity.)

The environmental implications of the PBR are in many ways similar to the other transmutor concepts. Thus, the impact of reduced demand for uranium ore on the risk to the public health is a positive attribute. In addition, there are the potential benefits from reduced actinide and fission product inventories in the long term. These are offset, however, by the increased risk to the public of accidents in reprocessing facilities and in transportation required for reprocessing.

State of Technology

BNL has no dedicated funding to support development of the burner PBR. However, the technology closely follows a reactor concept that BNL is developing for the Air Force. Some of the key technologies have also been demonstrated in the HTGR experience. This includes fuel-particle manufacturing (with plutonium content) and core-component frit manufacturing.

As proposed by its developers, the main areas of technology to be addressed in order to determine conceptual feasibility are

1. fabrication of fuel and target particles with different types of loading, such as MAs and fission products;
2. the capability of a fuel particle to retain fission products while being irradiated to the high desired burn-up and under the expected mechanical and thermal cycling conditions;
3. testing of several steps in the fuel and target materials reprocessing, including the electrolytic graphite dissolving method, as well as the scaling of the TRUEX process;
4. radiation damage effects (fluence of 10^{23} neutrons/cm^2) on core structural materials and moderator materials;
5. the uncertainty in thermal hydraulics due to nonuniformities in fuel loadings and fission power distribution; and
6. the fuel loading/unloading speed that allows for a rapid turnaround in the total inventory.

Other issues that need to be addressed include the delivery of high-speed helium coolant at a reasonable pressure drop and with high reliability. Also, the effects of burn-up reactivity swings on the operations of such a reactor remain to be evaluated, together with the power distribution issues characteristic of high thermal flux systems (also discussed for the ATW). In addition, the degradation of structural materials in high flux reactors (e.g., the high-flux isotope reactor) due to high energy neutrons and transmutation induced by neutron capture of alloying elements or impurities has been a major concern. This concern about degradation would probably have significant impact on the design of the PBR. Finally, the chemical processing of the fuels involved is a challenging problem that has to be resolved.

Cost Information

Development Costs

Detailed analysis of the cost of development is not available. BNL states that the development process will cost $1.2 billion and fall into the following stages:

Phase 1:	2 years -	$5 million to investigate basic feasibility issues.
Phase 2:	3 years -	$50 million for a detailed design and to test critical materials processes.
Phase 3:	3 years -	$150 million for engineering design and fuel-element nuclear testing.
Phase 4:	4 years -	$800 million to construct prototype reactor and particle processing facilities.
Phase 5:	2 years -	$200 million to test at operating conditions.

These costs appear not to include the developmental cost of the chemical separation processes.

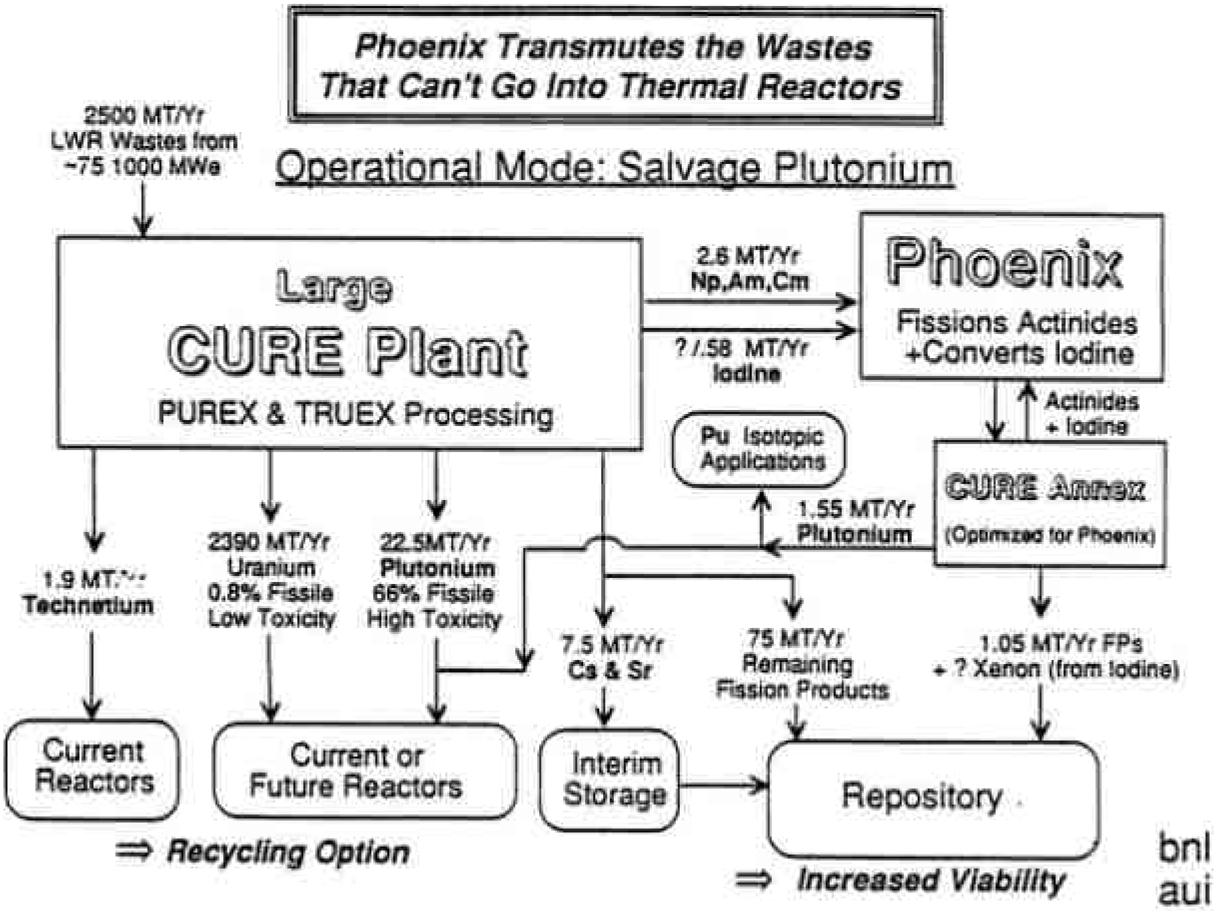
Systems Costs

Only rough estimates have been made for capital costs. The PBR (1,000 MWt) plant is assumed to cost $1 billion dollars on the basis of $3,000/kWe equivalent. An LWR PUREX/TRUEX plant with 1,500 MT/yr was estimated to cost $1.4 billion (i.e., about $1,000/MTHM). The PBR reprocessing plant with 250 MT/yr capacity was estimated to cost $1.2 billion. For 100 GWe of LWRs, given the support ratio of one PBR for five LWRs, the added cost of PBRs and separation facilities for waste processing can be $45 billion. If these cost estimates are accurate, this represents an investment equivalent to 15 to 20% of the power-producing LWRs alone. These cost estimates do not take into consideration the MOX fuel manufacturing facility that will be required. Alternatively, if all the plutonium is to be transmuted in the PBR, larger cores and higher costs will be associated with the PBR itself.

The Accelerator-Based Phoenix Concept

Reference Design

Phoenix is an accelerator-based transmuter concept being developed by Brookhaven National Laboratory (BNL). The transmuter is envisioned to be part of a larger scheme for separating and transmuting wastes from LWRs, as shown in Figure F-27. The objective for such a scheme, according to the proponents, is to reduce the time required for storage of nuclear wastes.



Source: Brookhaven National Laboratory

FIGURE F-27 Schematic flowchart of waste streams in Phoenix-based waste management network.

In this scheme, LWR waste is first separated into six streams: (1) technetium isotopes; (2) cesium and strontium isotopes; (3) MAs (neptunium, americium, curium isotopes); (4) iodine; (5) the remaining fission products; and (6) uranium and plutonium (as separate substreams).

The uranium, plutonium, and technetium would be recycled back into power reactors, thus requiring at a minimum the acceptability of an LWR-fuel recycle program. The cesium and strontium isotopes would be kept in interim storage (although this is really not an essential element of the scheme) and would ultimately be disposed of along with the residual fission products. The MAs and iodine (^{127}I and ^{129}I) would be burned in the Phoenix transmuter. Since the transmutation is incomplete and additional iodine, technitium, and fission products are produced during transmutation, a second separation facility would be required to separate the Phoenix waste into a similar set of waste streams for recycle, transmutation, and disposal (Figure F-27).

In the transmuter, an accelerated proton beam is used to both transmute MAs directly by spallation and to produce an intense flux of high energy (hard spectrum) neutrons to further transmute the MAs. The target assembly is maintained at a relatively high k_{eff} (less than 1) so that most of the neutrons come from neutron-induced fission rather than spallation (e.g., about 50:1). Since leakage of the neutrons out of the target is substantial—and provides some means of control of the assembly—the opportunity to moderate these neutrons and use them to burn iodine isotopes presents itself.

The inherent potential advantages of such a transmuter over some critical-reactor-based concepts (e.g., the advanced liquid-metal reactor [ALMR] and the LWR-based burner) are twofold: (1) the high-flux/hard-spectrum results in shorter times to achieve significant inventory reduction factors of the MAs vis-a-vis ALMRs (see Figure F-28), and (2) an accelerator-based neutron source has potentially improved safety features, as the assembly can be (in theory) maintained subcritical, and transmuter shutdown can be accomplished relatively quickly by switching off the proton beam. Decay heat and reactivity swings in response to temperature and power changes, however, would still be issues. Moreover, to realize their potential advantages, accelerator-based transmuters inherently add a relatively expensive (multibillion dollar) machine (accelerator) to an already expensive near-critical reactor (and separations scheme), and hence must generate additional revenue (e.g., electricity production) in a cost-effective manner to compensate for this expense. The specifics of these pros and cons are discussed below.

A schematic of the Phoenix transmuter is shown in Figure F-29. It is based on a linear accelerator producing a 104 mA beam of 1.6 GeV protons. This is not necessarily an optimal set of beam conditions, but a high energy is desirable for purposes of distributing the beam load through the target thickness, and as high a current as possible is desirable from the point of view of maximizing the neutron flux and hence the burn rate. In any case, these beam parameters are believed to be achievable. The knowledge base for evaluating the achievability of such a machine is identical to that for the LANL-accelerator transmutation of wastes (ATW) concepts, and so the difficulty in achieving this performance level is virtually identical, that is, development of such a machine is feasible but will require considerable engineering improvement over existing technology. Moreover, the accelerator technology is less ambitious than that envisioned for

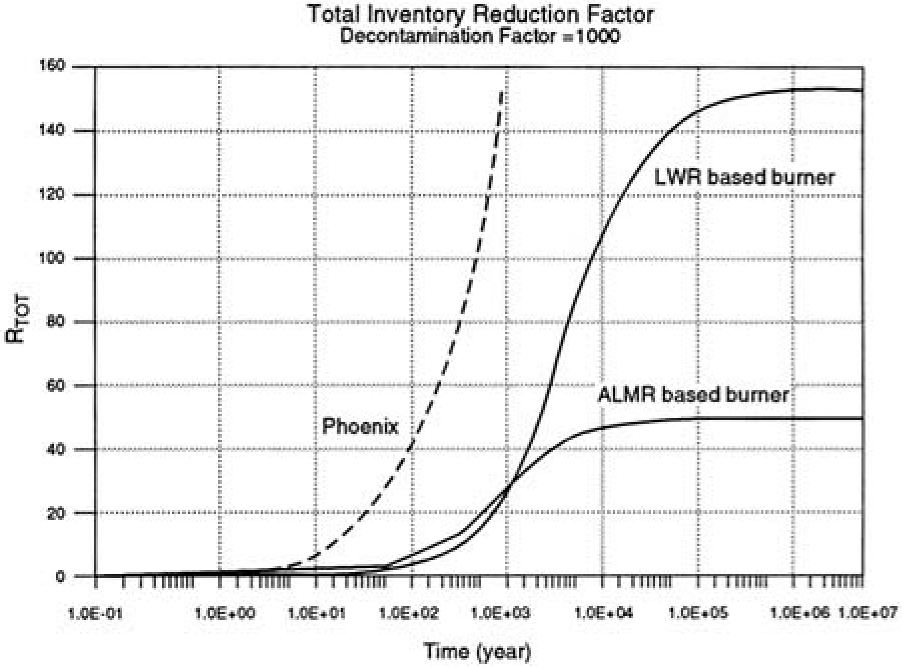
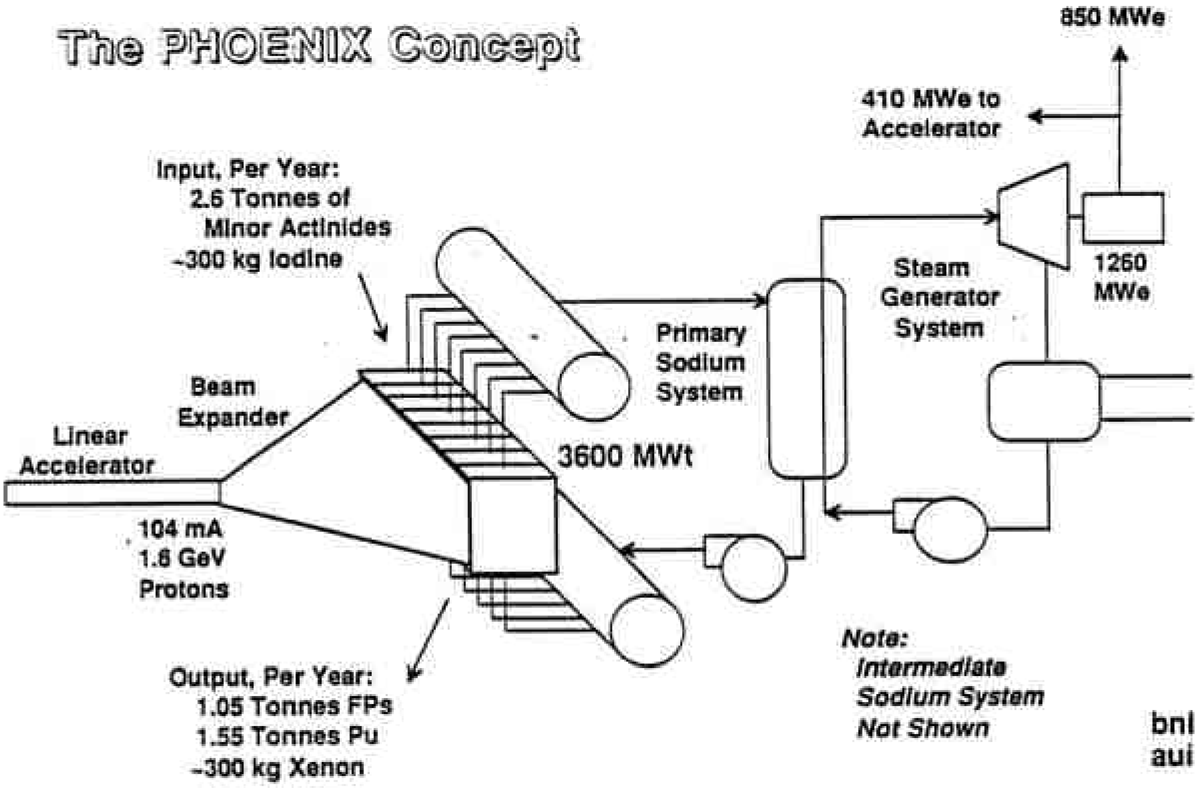


FIGURE F-28 Comparison of inventory reduction factor for the Phoenix and ALMR scenarios.



Source: Brookhaven National Laboratory

FIGURE F-29 Schematic of the Phoenix machine.

tritium production (Lawrence, 1990), and would not require funneling at the front end for the currents anticipated.

Unlike the ATW, in the Phoenix system the proton beam is spread (rastered) and impinges on an MA target directly. This produces spallation (fission and evaporation) and consequent high energy neutron production; the spallation neutrons induce subsequent fission (with attendant neutron production) in the MAs in the target. Since only 1 in about 50 neutrons comes from proton-induced rather than neutron-induced fission for a k_{eff} on the order of 0.9, the neutron spectrum will be dominated by the fission neutrons, and hence it is argued that it will be similar to that of a fast reactor with the same fuel/target type. However, in reality the neutron spectrum will still exhibit the high energy tail (i.e., neutrons with energies greater than 20 MeV) typical of spallation sources (albeit to a smaller extent), which may introduce some transmutation effects not considered in fast reactors (see below).

Since MAs are burned by a combination of spallation and neutron-induced fission, this combination improves the transmutation efficiency per proton and produces slightly more thermal energy per energy deposited by the proton beam (3,600 MW for 166 MW of beam power) compared with the ATW concept (3,600 MW for 200 MW of beam power). In addition, as noted above, by surrounding the target with an iodine-containing blanket and some moderator, the iodine can be transmuted to xenon isotopes by neutron absorption in the epithermal regime where capture is accelerated. Preliminary concepts assume a D_2O moderator in the iodine-containing blanket.

The MA target is envisioned to consist of a set of FFTF–like hexagonal fuel assemblies arranged in separate modules. The modular concept permits the accelerator to work on transmuting less than the total capacity of the machine. Eight modules (each .75 m high × .82 m × 1.88 m) would contain 124 hexagonal target assemblies (each 0.116 m across flat-to-flat and containing 217 fuel pins). These are similar to FFTF fuel designs. However, the Phoenix "fuel" initially contains only MAs and no plutonium, since plutonium is recycled back into existing reactors in this scheme; in part this is done to maximize the MA inventory while maintaining the system subcritical. However, plutonium builds up by radioactive decay of some of the MAs during operation and is removed periodically. Since no plutonium is in the initial fuel charge, there are insufficient neutrons produced in Phoenix to burn both technicium *and* iodine. This is the reason for separating technitium and recycling it to the power reactors for burning as well.

Although the concept is not currently tied to a particular fuel concept, an oxide-based fuel scheme is currently favored for reasons of experience with in-reactor performance and aqueous separations of oxide-based fuels compared with, for instance, a carbide or metallic fuel. Moreover, the system is currently being designed so that peak fluxes and neutron spectra are similar to the FFTF environment. Thus, although no detailed analyses of target performance have as yet been undertaken, first-order guesstimates are that fuel performance will be similar to FFTF oxide fuels and design burn-ups are currently less than peak burn-ups achievable with current oxide fuels. However, the actual experience base with purely MA-oxide fuels is virtually nonexistent in terms of in-reactor performance and aqueous separation. Moreover, the superposition of proton and neutron irradiation damage will need to be addressed for these target assemblies. Again the transmutant rates from a neutron spectrum with a high energy tail can be significantly different. This may be particularly important in the target cladding and structural components, where transmutant helium could degrade mechanical properties or induce void swelling at a faster rate than the fast breeder reactor experience to date. And spallation products of the light (oxygen) component of the oxide fuel or the fission products could introduce new chemistry effects not seen to date. Finally, even with the experience to date in aqueous processing, the postirradiation processing of these fuels will be challenging, as the isotopic make-up will be very different from fission-reactor fuels and will vary during the initial operation of the facility until a pseudo-equilibrium is achieved in the fuel cycle.

Again borrowing from liquid-metal reactor (LMR) experience, the target is to be cooled by liquid sodium, and the thermal power is to be used to generate 1,260 MWe of electrical power through an intermediate-heat-exchange loop and steam cycle. Of this 410 MWe would be returned to the accelerator and 850 MWe sold to the grid. Hence, the effective plant efficiency would be 24% (850 MWe/3,600MWt).

As noted earlier, the system requires both front-end processing of LWR spent fuel and intermediate processing of the Phoenix targets. The BNL group has not specifically worked on the separations scheme to date but has relied on analyses associated with the Clean Use of Reactor Energy (CURE) study referred to earlier in this report, and assumes a combination of PUREX/TRUEX to achieve separations of the six waste streams.

A 2-year fuel cycle has been assumed in nonoptimized scoping studies to date. (See flowsheet developed for Phoenix.) One Phoenix transmuter would accept 5,200 kg of MAs and 1,160 kg of iodine plus its own recycle (about 19,500 kg of MAs and recycle iodine) every 2 years. It would discharge 3,100 kg of plutonium every 2 years along with 1,160 kg of transmuted xenon (from iodine) and 2,100 kg of fission products for disposal. The iodine/xenon cycle has not been engineered for optimization, but it is currently assumed that iodine is burned at the same rate it is introduced, however, the total iodine inventory in the reactor remains to be worked out. Hence, one machine could burn MAs and iodine from 75 1,000-MWe LWRs on a steady-state basis. A second machine could burn stockpiled actinides from commercial spent fuel in—it is claimed—in about 40 years.

The initial charge to the transmuter is 24,700 kg of MAs. The MAs deplete while plutonium builds up. A short-lived curium isotope dictates the transient behavior of the curium inventory, but curium ultimately assumes a pseudo-equilibrium level after about 12 cycles of burning and recycling. The k_{eff} also increases substantially during a burn cycle; initial calculations indicate that it can swing from an initial value of about 0.8 to 1.05 at end of cycle, and so some additional control measures (increased leakage, introduction of poisons, etc.) will have to be taken to maintain the system subcritical. After 2 years, the total TRU inventory falls to about 22,500 kg, of which 3,100 kg is plutonium. This plutonium is largely (86%) ^{238}Pu, and proponents argue that there may be some opportunity to separate the ^{238}Pu for either radioisotopic power sources or "poisoning" weapons grade plutonium for proliferation control. In the Phoenix scheme, this plutonium is removed from the MAs discharged from Phoenix. The remaining (separated) 19,500 kg of neptunium, americium, and curium are combined with 5,200 kg from the LWR spent-fuel reprocessing to restore inventories to their initial levels upon reload. It is currently assumed (based on CURE) that reprocessing takes two years.

Transmutation Performance Requirements

As noted previously, the stated objective for such a scheme is "to reduce the time required for storage of nuclear wastes." Although not stating it explicitly as a goal for Phoenix, the BNL proponents have noted that partitioning losses of 10^{-5} or less should be a minimum objective for any partitioning/transmutation scheme if it is to succeed in the United States. The rationale for this position is that if partitioning losses of 10^{-5} or less can be achieved for each of the "problem isotopes" (defined as ^{237}Np, ^{241}Am, ^{243}Am, ^{244}Cm, ^{90}Sr, ^{99}Tc, ^{129}I, and ^{137}Cs), then NRC standard 40CFR191 (that specifies how many curies per ton of heavy metal of these isotopes can be allowed to escape the repository in the first 10,000 years following disposal) could be met without containment that ensures isolation of the waste. Moreover, in theory, HLW could be converted to a waste meeting radionuclide specifications of Class C waste according to standard 10CFR61 if this same level of portioning loss could be met (although it is recognized that standard 10CFR61 does not apply to HLW). Net partitioning losses of 10^{-5} require process decontamination factors of 10^6 or better on some elements (e.g., americium and curium) to achieve the specification for Class C waste with a 10% transmutation machine. This puts severe demands on the separations side of the scheme.

Facilities and Support Requirements

The scheme envisioned would require the following components to support one or more Phoenix irradiation facilities:

1. front-end reprocessing of LWR spent fuel, probably by a combination of PUREX and TRUEX, to separate waste into the requisite six streams with the process decontamination factors stated above (as noted above, this would have to operate with process decontamination factors of 10^6 or better, and; as in other schemes, effective decontamination of cladding hulls and process equipment would also be needed);
2. remote target fabrication for the MA and iodine targets for Phoenix and remote fuel fabrication for the plutonium and technetium recycle into power reactors;
3. intermediate reprocessing of Phoenix targets to separate plutonium and fission products from the MAs and iodine from the xenon products; again process decontamination factors of 10^6 would be required if the "minimum objective" for a transmuter is to be met by Phoenix; and
4. an interim storage facility for cesium and strontium, if these isotopes are to be separated from the other fission product wastes.

Residual Waste Management and Environmental Impact

The primary benefit of this scheme is to minimize the inventory of species (MAs, iodine, and technetium) in the waste repository that contribute most significantly to the current estimates of long-term risk. If separation and isolation of cesium and strontium isotopes are also accomplished, their removal from the waste repository would substantially reduce the near-term heat load on the repository and would permit a higher concentration of waste to be disposed of; this would postpone the need for repository expansion or addition. If the "minimum objective" is met, the partitioning and transmutation scheme would obviate the need for a HLW repository at all.

Risk and Safety Analysis

To date, no detailed risk and safety analyses have been made. Hence, at this point only some generic issues for transmuters in general, and accelerator-based transmuters in particular, can be anticipated.

Safety

The promoters point to the speed at which accelerator-driven sources can be "turned off" and to the inherent safety of a subcritical assembly (i.e., that supercriticality accidents would not be design-basis accidents). However, the large reactivity swings anticipated for a 2-year cycle ($\Delta k/k$ ` 0.25) and steps to counteract this (increased leakage, poisons) could introduce substantial changes in reactivity coefficients (e.g., sodium void worth, Doppler coefficients, etc.) that will have to be addressed; moreover, there may be safety issues associated with a failed beam raster (e.g., burning holes in targets with the attendant radioactivity release). In addition, decay heat will still be an issue, even with a rapid removal of the proton source. A liquid-sodium-cooled system provides an opportunity to configure the system for passive cooling under accident conditions, but this has not yet been addressed specifically.

Safety considerations for the balance of the plant are probably similar to those for LMRs in general (sodium fires, sodium–water reactions, etc.). Of somewhat unique concern is the potential for Na-D_2O reactions between the sodium-cooled MA targets and the D_2O-cooled and moderated iodine blanket.

Licensing

Licensing a Phoenix facility, a combination of a large accelerator and a near-critical fast reactor, can draw on issues and procedures developed for the component facilities to date, but new safety issues associated with operating the combination will have to be identified and resolved. Each of the support facilities (reprocessing, remote fuel fabrication, interim storage) has its own set of licensing issues that are not unlike those associated with these facilities for other transmutation schemes. The operational assumptions for this scheme are that the following activities are acceptable to the public and can be carried out in a manner acceptable to regulatory agencies:

- centralized fuel reprocessing;
- plutonium recycle in LWRs;
- the operation of a Phoenix-type waste transmuter;
- the operation of one or more reprocessing facilities for Phoenix waste;
- surface storage of separated radioisotopes for long periods (mainly cesium and strontium); and
- substantially increased cost of electricity (see below) to pay for (perhaps only perceived) long-term risk reduction in nuclear waste management.

Risk

Risk analyses will have to include:

- decreased risk from uranium-mining/milling as a result of plutonium recycle and energy generation from actinide burning;
- increased short-term risks associated with transmutation and separations operations and surface storage versus disposal of radioisotopes; and
- reduced long-term risks associated with inventory reductions of long-lived and water-soluble isotopes.

State of Technology

The Phoenix concept is still at a very preliminary stage. BNL has had no dedicated funding to support the development of Phoenix. Work has primarily proceeded using limited internal funding and a lot of voluntary effort.

While a team of competent people have examined the first-order issues, feasibility studies are still required to proceed. Outstanding technical issues include the following:

Accelerator performance: As noted earlier, the development group appears to have made reasonable estimates of what can be achieved in accelerator technology based on current knowledge, but considerable engineering improvements will be required to actually achieve this. These issues are common to ATW, although the accelerator requirements are less severe than for ATW-1.

MA target performance: Again, almost no analysis of target performance has been made, other than to assume that by designing a target environment similar to FFTF, no "show stopper" materials issues will be introduced. Under these circumstances, 2-year fuel lifetimes are probably reasonable. However, there is little experience with purely MA-oxide materials in such an environment, and the integral performance of an MA-oxide fuel in this environment remains to be demonstrated. Moreover, the combined effects of proton and neutron irradiation on structural materials will have to be addressed (although this is probably minor). Issues of the influence of transmutants introduced by high energy neutrons, along with the "usual" list of radiation-damaged induced property and dimensional changes (hardening, embrittlement, accelerated creep, void swelling), will have to be addressed for the specific materials and environments envisioned for the Phoenix targets.

Iodine transmutation: Little has been done to assess the iodine transmutation, other than to assume this can be done in a blanket surrounding the target. Further conceptual design is required; however, this is probably not a critical issue.

Separations: As noted, the work to date has relied on the CURE study for guidance in what might be feasible from a separations point of view. Hence, all of the outstanding technical issues for CURE apply here as well. These may be exacerbated, however, if process decontamination factors of 10^{-6} are required in full-scale reprocessing plants to meet the "minimum objective."

Additional waste: Additional waste generated by systems (contamination of fuel cladding hulls and separations equipment, radioactivation of Phoenix system components, etc.) has not been addressed. First-order estimates are that this will be small compared with the main sources of inventory. However, the impact of all activities on all of the waste streams needs to be addressed in any comprehensive incremental (decremental) risk analysis, as discussed above.

Costs

Development Costs

Based on numbers provided by BNL and figures from similar proposals, a development effort is estimated to consist of the following:

- *Phase I:* a conceptual design study to address the issues listed above; the study would take at least 2 years at a funding level of about $12.5 million per year;
- *Phase II:* an engineering design study combined with the development of an engineering database for critical components (accelerator, target, balance of plant); this would be about an 8- to 10-year study at a level of about $100 million per year;
- *Phase III:* construction and operation of a demonstration plant; the cost may be near $2 billion and would take about 5 to 8 years to construct and demonstrate operations.

Phases I, II, and III would overlap.

Systems Costs And Economics

Again, the estimates here are based on preliminary input. The estimated cost of the accelerator is about $2 billion (1992 dollars). The estimated cost of the subcritical target, sodium loops, and power plant is $5 billion (for a large power plant). The operation and maintenance costs are expected to be double that of a large power plant.

As noted above, the effective efficiency of the plant would be about 24%. Coupling this with the 40% increase in the capital cost and a doubling of the operation and maintenance costs would mean that electricity would cost roughly twice as much to produce in Phoenix as in a large reactor. Hence, the Phoenix operator would either have to find a buyer for this expensive electricity or, more likely, charge the nuclear industry the difference in the name of improving waste disposal. If the average cost of production were about 7 cents/kW·h, this would mean charging the industry about $420 million per year in addition to separations and disposal costs to annually burn 2,600 kg of MAs (and 580 kg of iodine), or about $132,000 kg of MA and iodine burned. This cost must be compared with that of burning MAs and iodine in LWRs or ALMRs, and evaluated against the "intrinsic safety" of such a machine and the speed of inventory reduction over the reactor-based systems.

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Appendix G

Effects on Repository

Background Information On Repository Performance

Repository Design and Operation

Nuclear waste repositories have been investigated since the late 1960s. Site evaluation and conceptual designs have been developed in many countries, including the United States, Sweden, and Germany. All concepts are largely based on disposal in repositories approximately 200 to 1,000 meters below the surface. The host rock for repositories varies. In the United States, volcanic tuff in the unsaturated zone at Yucca Mountain is now the only rock type being investigated. In Germany, a salt dome is being characterized, while Sweden is concentrating on granite under saturated groundwater conditions as the primary focus.

An underground configuration for the proposed Yucca Mountain repository, shown in Figure G-1 (reference: Yucca Mountain Site Characterization Plan), is representative of that in most repositories. Waste packages containing either spent fuel or solidified waste forms from reprocessing are emplaced in the floor of the mine drifts or, in some cases, directly in the drifts. In the United States, the wastes will consist of spent fuel removed from the reactors and some solidified high-level waste from U.S. defense activities. Each package will contain a few metric tons of spent fuel or an amount of reprocessing waste approximately equal in radioactivity and thermal power. Approximately 40,000 waste packages would be distributed over an area ranging from 1,000 to 2,000 acres. In the United States, each waste package is expected to include a high-integrity container capable of providing up to 1,000 years of total containment. The current design for the Yucca Mountain repository allows for a total of 70,000 metric tons uranium (MTU) with 63,000 MTU of spent fuel. Higher loading densities or use of additional area would allow an even greater capacity.

The operations at a repository consist of a major facility for receipt and packaging of the waste and underground excavation for waste emplacement. After waste is prepared in special canisters, it is moved underground and emplaced in a manner to preclude radiation exposure to operating personnel. The annual capacity of the repository is determined by the ability to receive, package, and emplace the waste. At Yucca Mountain, an annual capacity of 3,400 MTU per year is anticipated after an initial startup period of a few years. The start-up operations are planned for 2010 and should continue for about 22 years.

The density of waste per unit area of a repository is primarily determined by the heat produced by the waste. Other factors that contribute are the allowable excavation patterns, permissible canister spacing, and the need to avoid certain geologic features. Typical waste forms produce approximately 1 kW of heat per MTU for fuel 10 years out of the reactor. However, by the time of the earliest possible repository operation in the United States,

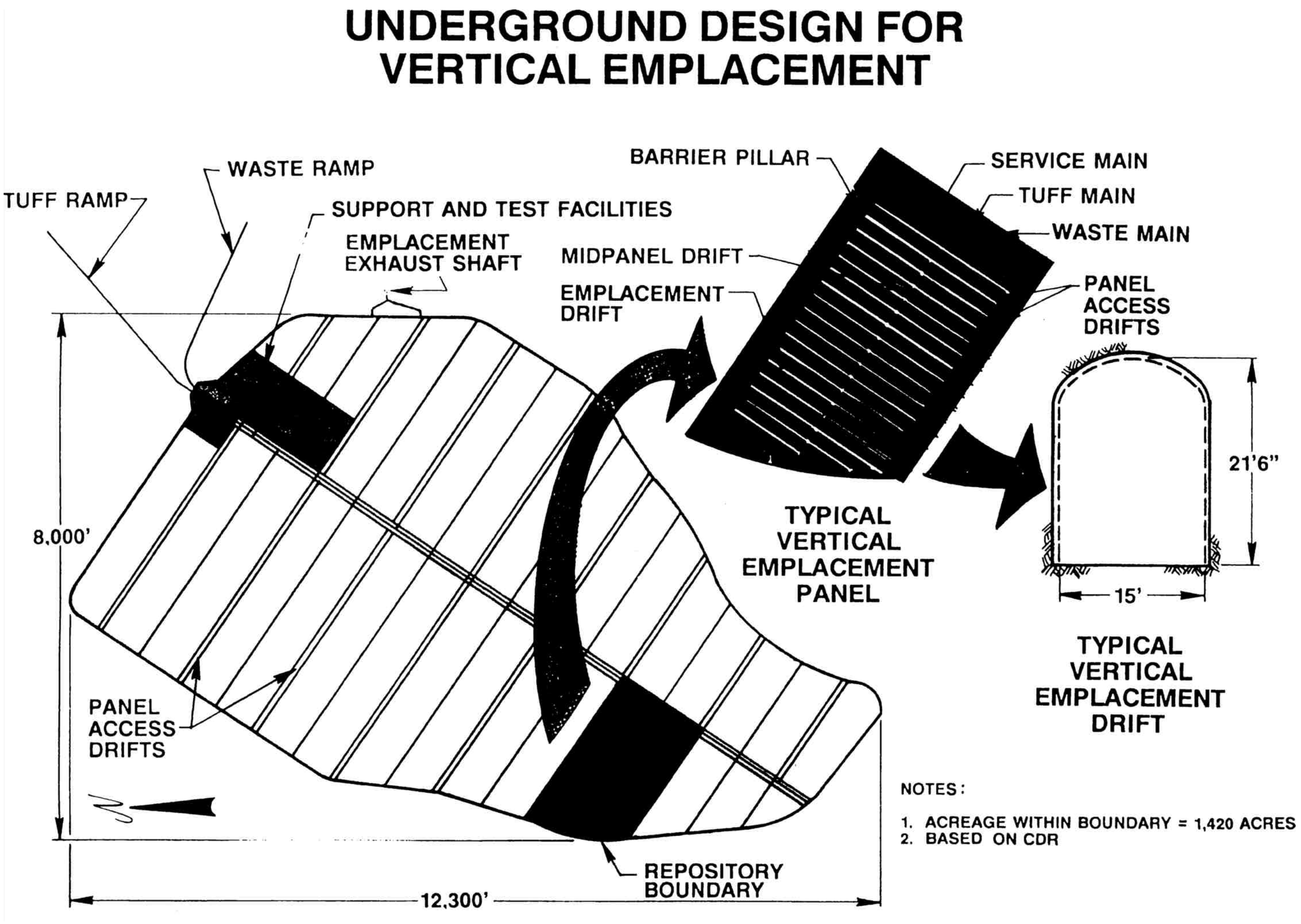

FIGURE G-1 An underground configuration for a nuclear waste repository.
SOURCE: Hunter et al. (1989)

approximately 60,000 tons of spent fuel will have been discharged with an average age of approximately 16 years. At least 10,000 MTU will have an average age greater than 30 years. The conceptual designs for repositories determine a reference value for thermal loading density by evaluating temperatures expected for the waste packages, the excavation drifts, and surrounding rock units. These predictions of temperature are compared with the expected operational and long-term performance concerns, and a loading density is established. At Yucca Mountain, approximately 57 kW/acre has been the reference value, although both higher and lower values have been considered. Waste emplacement configurations and schedules can be designed to achieve a wide variety of heat loading patterns, especially if underground ventilation is used to remove heat during operation.

Long-Term Performance Assessment

Basis For Evaluation

The standards for the necessary resolution of radioactivity in nuclear waste repositories are still being developed; however, all proposed standards consider limits on the potential dose to individuals, total population doses, or maximum radioactivity releasable over periods of time 10,000 years and longer. Repository developers must evaluate all possible mechanisms that might result in releases of radioactivity to the environment and must determine the risk to humans that is associated with this release.

Three measures that have been adopted or proposed in various countries to evaluate potential public health risk from geological disposal of radioactive wastes are:

1. annual radiation dose to an individual,[1] which is usually applied for expected releases or high-probability events;
2. annual risk to an individual from radiation exposure, which takes into account the probability of exposure; and
3. collective radiation dose to populations, integrated over all population exposed and over time from the beginning of geologic disposal.

Possible benefits of partitioning and transmutation relative to each of these performance measures are discussed herein and are based on published analyses of the performance of conceptual repositories in various geologic media.

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[1] It is the practice in some countries to calculate the dose to the maximally exposed individual, a person whose entire intake of water is contaminated by radionuclides released from the repository and whose entire intake of food is grown in or nurtured by the contaminated water. Other analyses focus on the doses to an individual in the critical population group, as recommended by the International Committee on Radiation Protection.

Approach To Evaluate Performance

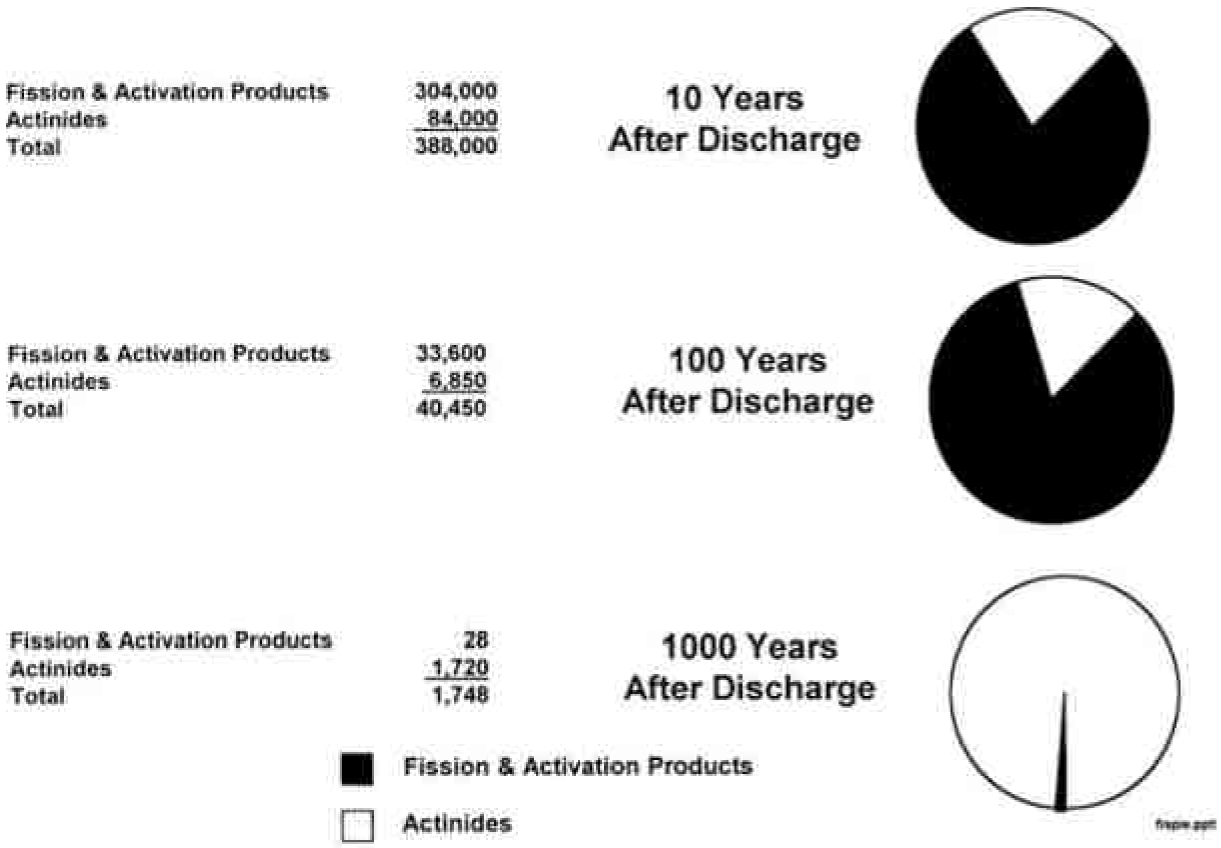
The U.S. Environmental Protection Agency (EPA) has established a derived set of limits on the cumulative amount of radionuclides that could be released to the environment over 10,000 years (EPA, 1985). These limits were derived from the EPA goal of no more than 1,000 health effects in 10,000 years for all the population exposed. The derivation assumed (1) that the radionuclides released are distributed throughout the surface waters of the world, (2) a constant population of 10 billion people, and (3) eating and drinking habits remain the same as today. Thus, the EPA limits are derived from a 10,000-year limit on collective dose, assuming that all repositories release radionuclides to the same environment as the EPA "generic" assumptions.

The U.S. Congress decided in 1992 that the current EPA standard shall not apply to the proposed repository at Yucca Mountain. A committee of the National Research Council is charged with evaluating and proposing a technical basis for a new standard for geologic disposal of high-level waste (HLW), such as a limit on individual doses, to be considered subsequently by EPA in promulgating a new standard. It is possible that a new standard may preserve some form of the EPA release limits, so the effects of separation and transmutation (S&T) on cumulative releases are included in the following discussion.

Long-term performance assessments describe two broad types of scenarios to assess the probability and quantities of radioactivity that might be released. The first includes evaluating all those processes that are reasonably expected to occur in the region of the repository. Principal among these are the "dissolution-and-migration" scenarios in which groundwater eventually penetrates the waste packages and slowly moves radioactivity to the accessible environment. In the unsaturated zone, it is also necessary to consider the movement of air in the host rock and the potential to transport gaseous radionuclides (e.g. ^{14}C in carbon dioxide. In most repository settings, these scenarios typically consider groundwater travel times to the accessible environment that exceed 1,000 years and most often 10,000 years. Further, they include an evaluation of the waste package release and migration potential of individual radioactivity in various chemical forms.

The second type of scenario includes "disturbed" conditions that might be expected to accompany undesirable geologic events such as earthquakes, volcanos, and abrupt changes in local or regional hydrologic conditions. These scenarios also include the effect of "human intrusion," in which people in future generations unknowingly penetrate into a repository and release a portion of its contaminants into the earth's surface or the groundwater system.

All performance assessments begin with assumptions about the form, characteristics, and radioactivity content of the wastes. The initial inventory for spent fuel consists of actinides, fission products, and activation products. The radioactivity content is distributed between the major isotopes, as shown in Figure G-2. Initially the short-lived fission products (^{90}Sr, ^{137}Cs) and a few short-lived transuranic (TRU) elements (^{241}Pu, ^{258}Pu, ^{244}Cm) dominate the radioactivity. However, for longer-term scenarios (greater than 1,000 years), only long-lived fission products (^{129}I, ^{135}Cs, ^{126}Sn, ^{99}Tc, and ^{79}Se) and certain actinides (^{243}Am, ^{239}Pu, ^{257}Np, ^{241}Am, ^{240}Pu, and ^{234}U) have a potential to contribute to releases. For gaseous releases, ^{14}C is expected to be the primary contributor to release.



SOURCE: Roddy et al. (1986).

FIGURE G-2 Contributions to total radioactivity content of spent fuel (Ci/MTU).

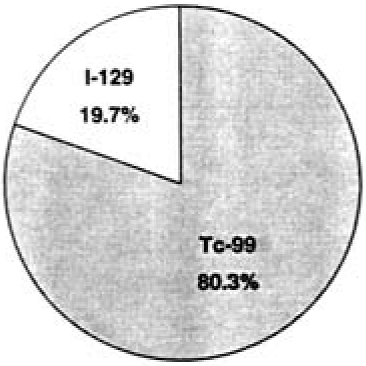
The predicted releases from dissolution-and-migration scenarios will be determined by radionuclides with long half-lives, high solubility in groundwater, and low sorption along the transport pathway. In unsaturated conditions like Yucca Mountain, the actinides have low solubility and high sorption, and thus less mobility, compared with the fission products; consequently, as shown in Figure G-3, only ^{129}I and ^{99}Tc contribute significantly to the releases to the environment. Further, these releases are typically shown to be much less than allowed by the original EPA standard. This is reinforced by the fact that these two radionuclides are only present in the initial inventory at concentrations below or equivalent to that allowed to be released under the EPA standard. The most notable exception to this general rule is ^{14}C, which contributes many times more to the release than any transport through groundwater. The unique nature of ^{14}C for an unsaturated repository deserves special attention for standards and regulatory development.

For "disturbed" scenarios, principally those involving human intrusion, radioactivity can be transported directly to the environment via the breaching or drilling operations. These events are assumed to occur over the history of the repository including some at very early times (less than 1,000 years). Thus sorption and half-life and, for the most part, solubility are not major factors in determining the potential release. Figure G-4 shows a typical distribution of contributions to the result of a human intrusion that involves release of radioactivity to the surface. In this case, the dominant contribution is from TRU elements (^{240}Pu, ^{239}Pu, ^{241}Am) with some contributions from fission products (^{137}Cs). Again, the total releases are small compared with those allowed by EPA standards.

Impact of Transmutation

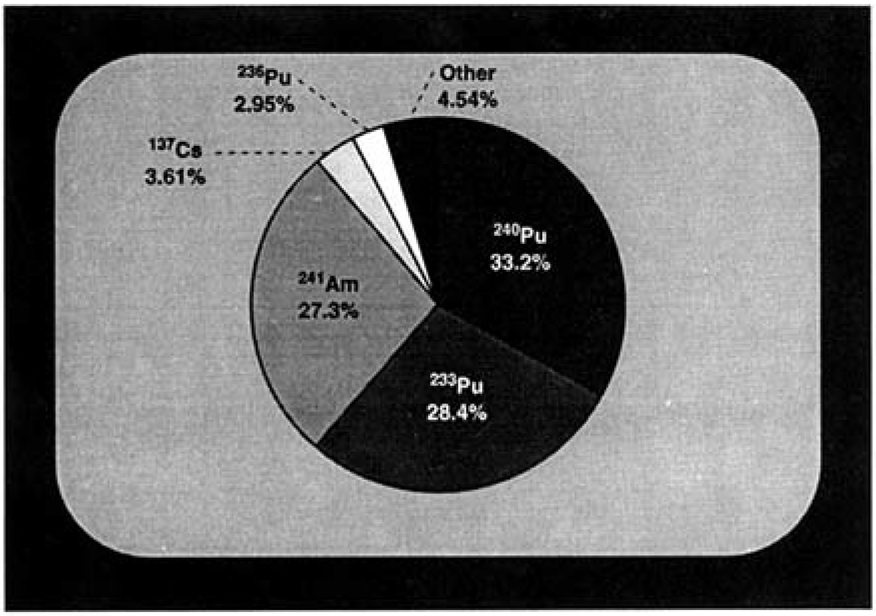
Thermal Effects

Partitioning transmutation significantly alters the radionuclide composition of the wastes emplaced in the repository. As a result, all temperature-dependent aspects of the repository are affected to varying degrees. This section describes and evaluates these effects. The analysis will focus on the effects resulting from S&T of the light water reactor (LWR) spent fuel (hereafter spent fuel) that is scheduled for emplacement in the first repository. There is essentially no difference between the various cases being considered because only one evaluation is required to characterize the evaluation cases considered in this appendix. Consideration of HLW from various fuel types would not be expected to change the qualitative aspects of the analysis or the conclusions, because the HLW essentially contains only fission products, which vary little in the amount generated per unit electricity generated. The discussion in this section emphasizes the Yucca Mountain study site as the potential location for the first repository because of the wealth of information already available on this site. However, the consequences of thermal effects on a saturated repository site (such as granite) are also discussed.



SOURCE: Barnard et al. (1992)

FIGURE G-3 Contributions of individual radionuclides to releases calculated using composite-porosity flow model.



SOURCE: Barnard et al. (1993)

FIGURE G-4 Contribution of individual radionuclides to releases on the surface from human intrusion.

Effect Of S&T On Thermal Power Of Waste

The decay of any radionuclide results in the emission of radioactive particles. These emanations are absorbed in surrounding materials and manifest themselves as heat. In the case of spent LWR fuels, the emanations are sufficient to require engineering measures (e.g., limits on the size of packages, active cooling measures, minimum package spacing) to keep the fuel within prescribed temperature limits.

The thermal-power profiles of spent LWR fuel and its various major constituent groups are shown in Figure G-5. After about 30 or 40 years, the thermal power of spent fuel comes from the short-lived radionuclides (^{90}Sr and ^{137}Cs) and the actinides (primarily americium and plutonium). The former decay with a half-life of about 30 years. The latter decay with a thermal-power half-life ranging from 400 to 500 years. The contribution of the actinides to thermal power equals that of the fission products after about 70 years out of the reactor and exceeds that of the fission products thereafter.

There are two S&T options that might significantly alter the thermal-power profile of high-level wastes (HLW) generated by processing spent fuel. Option 1 involves removal and destruction of the TRU actinides (uranium does not contribute significantly to thermal power), which results in the generation of actinide-free HLW. The amount by which the thermal power is reduced is shown in Table G-1 for selected decay times.

TABLE G-1 Relative Thermal Power of Actinide-Free HLW and Spent Fuel

Decay Time (years)	10	30	100	300	1,000
Thermal Power (percent) Ratio[a]	83	70	34	0.8	0.001

[a] Thermal power of spent fuel without actinides to unprocessed spent fuel.

Option 2 involves removal of the TRU actinides plus ^{90}Sr and ^{137}Cs. In this case, the thermal power is reduced to low levels (about 1% of the spent-fuel thermal power at 10 years) and declines steadily thereafter.

Consequences Of Altered Waste Thermal Attributes To Repository Capacity

Removal of major constituent groups from spent fuel as a part of processing could significantly increase the amount of waste that can be emplaced in a unit area of a repository

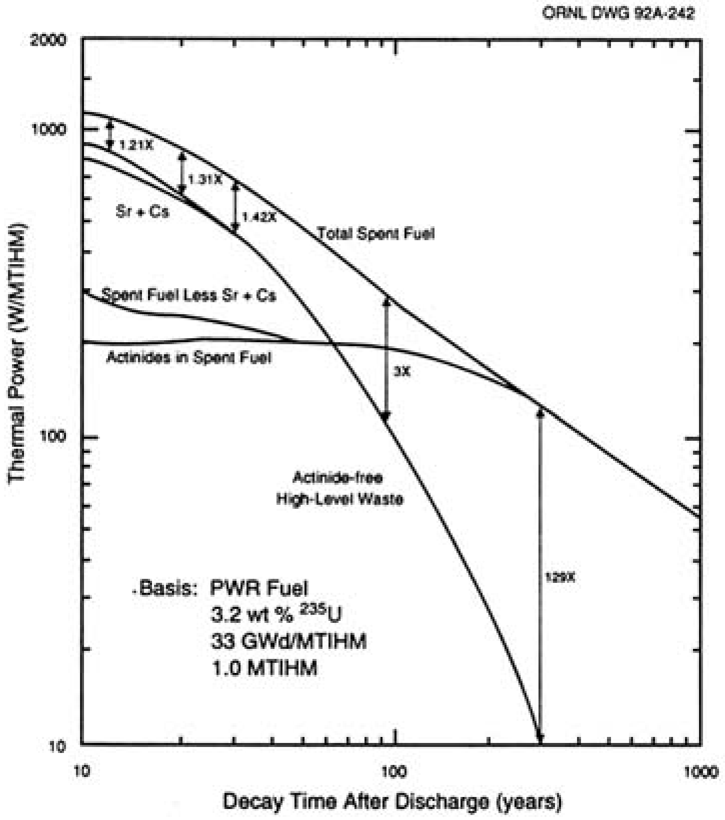


FIGURE G-5 Thermal power profiles of spent LWR fuel and its major constituents groups.

site. This increase is directly related to the total capacity of a finite repository site, and thus, to the time during which it can receive and emplace HLW.

To the extent that these consequences have been studied (mostly scoping studies), they appear to be qualitatively the same for both saturated and unsaturated repository sites. As a result, the following discussion does not attempt any differentiation between these sites.

Option 1. Actinide-free HLW differs in thermal attributes from spent fuel in two distinct ways that may increase the capacity of the repository. The first is that removal of the actinides immediately reduces the thermal power of the waste from a unit amount of power generation by about 20%. Because the spacing of waste packages is limited by the temperature limits of the waste form (e.g., spent-fuel cladding, borosilicate glass centerline), the reduction in thermal power would allow about 20% more of a given waste form to be emplaced in a given area.

The second change in thermal attributes is that the actinide-free HLW thermal power declines much more rapidly than for spent fuel. It is possible to design waste-emplacement scenarios in which the waste is initially emplaced relatively far from its nearest neighbor and meets applicable limits. Additional emplacement drifts are constructed (currently, drift spacing is limited by thermal considerations), and subsequent HLW packages are emplaced in these drifts. After about 60 years of emplacement, continuation would require that the next package be emplaced between the first two packages emplaced in the repository. Because of the short half-life of strontium and cesium, the thermal power of the first two packages would only be 25% of the initial thermal power, and this should allow applicable limits to be met. This approach would continue until new packages were emplaced between the early-emplaced, widely spaced packages.

Scoping calculations indicate that, taken together, the above effects would allow the capacity of a unit amount of repository area to be increased by factor of 4 to 5 (assuming about the same receipt rates as those projected for the Yucca Mountain study site) as compared with a relatively aggressive spent-fuel emplacement case specified by Johnson (1991). It should be noted that this conclusion would not apply to the earlier concepts of ATW cases or other cases in which the thermal efficiency deviates substantially from that of LWRs and liquid-metal reactors (LMRs).

Although the same technique can be employed to a limited extent with spent fuel, the benefits are thought to be minimal (perhaps a 20% increase). This occurs because the thermal power declines much more slowly with actinides present, and other temperature limits specified in the site characterization plan of the Yucca Mountain site are soon encountered.

In the case of both spent fuel and HLW, a number of other measures might be taken to increase the capacity of a unit amount of repository (Ramspott, 1991). These might include fuel/waste aging, devices to enhance heat transfer from the waste package into the emplacement drift, and reevaluation of thermal criteria. In general, these measures would be complementary to the capacity benefits of actinide removal.

Such an approach is not without penalities, which include a more complex emplacement scenario, the need to maintain mined openings for very long times, and the increased and continuing need for ventilation to remove the heat. These disadvantages must be weighed against a capacity increase that would greatly delay the need for another repository and the controversial and expensive activities that accompany it.

Option 2. In this option, ^{90}Sr, ^{137}Cs, and the actinides are removed and the thermal power of the waste is reduced to very low levels as compared with spent fuel. This means that the waste form temperature limits of the waste become essentially irrelevant, which yields the following benefits if the waste continues to be emplaced according to the concept specified in the site characterization plan (e.g., in vertical boreholes): (1) the diameter of a waste package can be much larger, which saves in package and handling costs; and (2) the waste packages can be spaced as close together as rock integrity considerations will allow.

The amount by which repository capacity might be increased in this option has not been calculated, but a factor of 5 should be approximately correct.

If alternative emplacement concepts are adopted, much larger capacity increases may be possible. The thermal-power level should be low enough to allow the wastes to be closely stacked in the drifts in lightly shielded packages or by shielded emplacement vehicles. Again, the magnitude of the capacity increase cannot be precisely quantified. However, factors of 10 to 40 appear to bound the possibilities.

This option entails one major penalty: disposition of the recovered strontium and cesium. The nuclear properties of ^{90}Sr and ^{137}Cs make them very difficult, if not impossible, to transmute. Because of the considerable emission of penetrating radiation and heat, they are clearly unacceptable for near-surface disposal. Thus, this material would presumably have to be stored in an engineered surface facility until it reached concentrations approximating the levels that come under the category of Class C LLW defined in the Nuclear Regulatory Commission (NRC) standard 10CFR161. This time ranges from about 200 to 500 years, depending on the initial concentration of the contained radionuclides. The radiocesium also contains a small amount of long-lived (3-million-year half-life) ^{135}Cs. The concentration of ^{135}Cs appears to be less than the limit for Class C LLW at all times, although definitive regulations have not yet been established. To be more precise, the penalty is the need to site, license, build, operate/monitor over an extended time and decontamination and decommissioning a major storage facility that is likely to be contentious. The facility might be much like the currently proposed monitored retrievable system (MRS) in most ways.

Consequences Of Altered Waste Thermal Attributes To Long-Term Repository Performance

Amount and duration of the thermal pulse from HLW as compared with that from spent fuel has a major effect on the physicochemical processes that occur after the repository is closed. Elucidation of these effects and their consequences is a very complex task that requires an extensive base of assumptions (e.g., emplacement scenario and waste characteristics), computational results (e.g., determination of temperature as a function of space and time), and site response functions (e.g., behavior of natural and engineered materials as a function of temperature and time). Because this information base is not yet available, the following discussion is based on broader generalities.

Saturated Sites. At typical repository depths (300 to 1,000 m), the water pressure is sufficiently high that the repository horizon will resaturate and remain saturated in perpetuity. After the repository is completely closed, it is typically assumed that it will resaturate over times that could range from immediately to several centuries. At the same time, the host rock is heated to a temperature much higher than ambient. Temperature maxima within waste packages (very near field) are expected within a few to several years after closure, based on heat conduction to surrounding rock, although this time might be extended if boiling occurred during repressurization. Maxima within meters of the waste package (near field) are expected within decades, and within centuries at distances of 50 to 100 m from the emplacement level (the far field). Thereafter, temperatures slowly fall as the thermal power of the waste declines exponentially.

This temperature pulse can potentially have a number of deleterious effects on the performance of repository components. Some of the most important impacts are as follows:

- The rate at which reactions (e.g., corrosion) occur in engineered materials typically increases exponentially with temperature.
- Radionuclide solubilities typically increase as temperature increases, although some of the actinides may exhibit retrograde solubility under some geochemical circumstances.
- Increased temperature can irreversibly degrade the ability of natural ion exchange materials (e.g., zeolites) to retard migrating radionuclides.
- The thermomechanical stresses induced in the surrounding rock can provide additional or more-transmissive pathways for water to enter repository, degrade waste packages, dissolve radionuclides, and transport them to the environment.
- Temperature differences can provide a driving force for movement of water through the repository horizon because of buoyancy effects.

As a result, in saturated repository, performance is expected to be affected as the magnitude of the temperature pulse increases. It then follows that any option that might reduce the temperature pulse should be beneficial to repository performance unless radionuclides exhibiting retrograde solubility were found to dominate repository risk, which is unlikely.

In the case of Option 1, the duration of the temperature pulse would be substantially reduced by removing the actinides. In approximate terms, removal of the actinides reduces the total amount of heat emitted by the waste by about a factor of 4 over the long term, allowing the duration of the elevated temperatures in the repository to be reduced. In the case of 30-year-old fuel, the time during which the repository center temperature is above 100° C is reduced from about 1,500 years to zero. Reduction of the maximum temperature could also be achieved but at the expense of repository capacity increases.

In the case of Option 2, which involves removing both actinides and strontium, the temperature pulse is essentially nonexistent, and thermal impacts would be expected to be essentially nil.

Unsaturated Sites. In an unsaturated site, the waste is emplaced above the water table. This is projected to be the case at the proposed Yucca Mountain study site. In such a situation, all of the disadvantageous effects of the thermal pulse, which are listed above, still apply.

However, in the case of unsaturated sites, there may be major advantageous effects of the thermal pulse. To understand this, one must first understand that the primary (although not only) radionuclide release mechanisms require that groundwater contact the waste package, degrade the package to expose radionuclides, dissolve the radionuclides, and transport them to the biosphere. The advantage of the thermal pulse is a result of designing the repository to prevent water from contacting the packages by keeping them hotter than the boiling point of water for an extended time. Although design calculations are not complete, it appears that it may be possible to maintain a dry environment at the surface of waste containers for many thousands of years. Ramspott (1991) has proposed an "extended dry" concept whereby the thermal pulse could be extended to maintain a dry environment for over 10,000 years.

The thesis that a "hot," unsaturated repository is preferable is not universally accepted. Some investigators (Eriksson, 1991) have concluded that the disadvantages of a larger temperature pulse outweigh the advantages and that a "cold" repository is preferable. This is based on the notion that a hot repository and significant changes in repository temperatures result in complex processes and events (human-induced perturbations) that are difficult to measure and predict and that are associated with significant uncertainty in safety assessments.

At this juncture, the committee endorses the National Waste Technical Review Board's (1992) views that "… a *technical basis* for the DOE's [Department of Energy's] current thermal loading strategy for the Yucca Mountain site does not exist" and that "… DOE needs to thoroughly investigate alternative thermal loading strategies. …" The primary reasons for this are the lack of a system-wide evaluation of the benefits and penalties of various thermal loading strategies, especially as they affect long-term repository performance and the fact that the current reference strategy is based on calculations that still need to be experimentally validated.

Assuming, for the sake of discussion, that a "hot," unsaturated repository is preferable, then removal of the actinides (Option 1) would clearly diminish the benefits of this approach, and removal of the actinides and strontium and cesium would completely eliminate these benefits.

Long Term Performance Effects

Important Radionuclides Of The Reference Once-Through Uranium Fuel Cycle Impacting Water Pathways And Individual Dose

To evaluate possible benefits from transmuting various radionuclides, it is necessary to first identify those species that are calculated to be the main contributors to long-term individual doses from geologic disposal. For this purpose it is sufficient to focus on the relative values of maximum annual dose to individuals from each radionuclide. This can be calculated by using relative values of annual doses and published information from other countries that present future dose rates from assumed failure of a single waste container. Relative values for the once-through uranium fuel cycle, resulting from dissolution in groundwater and transport to the biosphere and calculated from published dose estimates for conceptual repositories in granite, are listed in Table G-2. The relative dose rate is the ratio of the maximum dose rate for a future individual and a given radionuclide to the maximum dose rate from the radionuclide that is the greatest contributor to future doses to individuals. Each maximum dose rate is calculated at the future time when that maximum occurs.

Estimated relative doses to future individuals from conceptual repositories in unsaturated tuff, loaded with spent fuel from LWRs, differ from those listed in Table G-2. The groundwater in unsaturated tuff is expected to be saturated with oxygen, resulting in an oxidizing atmosphere surrounding the waste containers. Containers are expected to fail by localized penetrations and cracks, allowing water to leak in and dissolve the waste solid. If the water within the waste container is as oxidizing as that in the surroundings, the solubilities of many important radioelements are expected to be greater than those that would occur in the reducing environment of a granite repository. The fission product ^{99}Tc would become much more soluble and is calculated to result in peak doses in the same range as those calculated for ^{129}I (Eslinger et al., 1993; Hirschfelder et al., 1991, 1992; Wilson et al., 1994; Andrews et al., 1994; Duguid et al., 1994). In a granite repository, the reducing environment provides sufficiently low solubility that the ^{99}Tc peak dose is well below that of ^{129}I, as shown in Table G-2.

However, it is not clear that the conditions within a waste container in unsaturated tuff will be oxidizing. If the bulk of the container remains intact after penetration, the container metal can be expected to promote a reducing environment within the container, particularly if the container is composed of thick metallic iron. If so, ^{129}I would remain the main contributor to peak dose, as shown in Table G-3.

The solubility of the actinide neptunium is particularly sensitive to the oxidizing potential, as well as to pH. Recent performance assessments for waste in unsaturated tuff (Wilson et al., 1994; Andrews et al., 1994; Duguid et al., 1994) have conservatively adopted a log-normal or log-beta distribution of neptunium solubilities ranging from 10^{-8} to 10^{-2} M. The assumed high solubilities result from the assumption that sufficient air-saturated groundwater enters through penetrations in failed waste packages in sufficient quantity to control the oxidation potential within the waste package. The effect of the metallic structure of the waste container to cause a reducing environment within the waste package has been neglected.

With such assumed distributions of neptunium solubility that emphasize the high end of the solubility range, the resulting peak doses of ^{237}Np can become greater than that of ^{129}I and ^{99}Tc. The peak doses of ^{237}Np are predicted to occur in the era of about 800,000 years. If the effective solubility of neptunium within the waste package does turn out to be near the high end of that assumed range, then either transmutation of ^{237}Np or improved waste forms containing separated ^{237}Np could improve the performance of a tuff repository.

The high individual doses appearing in the recent studies of a repository in unsaturated tuff are a consequence, in part, of the simplifying assumption that a failed waste container would present no barrier to the release of radionuclides from a waste package. If the container fails by localized small penetrations, as is expected by the designers, the small penetrations can present a significant impedance to the transport of groundwater through the container wall, thus reducing the effect of aerated groundwater on the oxidation potential within the container.

TABLE G-2 Relative Doses to Future Individuals From a Granite Repository Loaded With Unreprocessed LWR Spent Fuel

		Relative Dose Rate[a]			
Radionuclide	Half-Life, (years)	Sweden-SKI[b]	Sweden-SKB[c]	Finland[d]	United Kingdom-NRPB[e]
^{129}I	1.57×10^{7}	1	1	1	1
^{99}Tc	2.11×10^{5}	1×10^{-5}		6×10^{-5}	6×10^{-1}
^{135}Cs	2.3×10^{6}	3×10^{-2}	1×10^{-2}	1×10^{-2}	1×10^{-1}
^{14}C	5.73×10^{3}	3×10^{-5}		6×10^{-2}	4×10^{-12}
^{36}Cl	3.01×10^{5}			2×10^{-3}	2×10^{-9}
^{79}Se	6.5×10^{4}	3×10^{-3}		1×10^{-2}	2×10^{-11}
^{126}Sn	1.0×10^{5}		1×10^{-2}	6×10^{-4}	2×10^{-14}
^{226}Ra	1.6×10^{3}		1×10^{-3}	2×10^{-2}	
^{231}Pa	3.28×10^{4}	2×10^{-2}	3×10^{-2}	4×10^{-1}	
^{234}U	2.45×10^{5}	1×10^{-8}		6×10^{-5}	
^{238}U	4.47×10^{9}	1×10^{-8}		1×10^{-5}	
^{237}Np	2.14×10^{6}	3×10^{-5}		4×10^{-4}	
^{239}Pu	2.41×10^{4}	3×10^{-5}		2×10^{-3}	
^{240}Pu	6.56×10^{3}	1×10^{-8}			
^{242}Pu	3.73×10^{5}	3×10^{-5}		4×10^{-2}	
^{243}Am	7.38×10^{3}			4×10^{-4}	

[a] Maximum dose rate from the listed radionuclide divided by the maximum dose rate from the radionuclide that contributes the largest dose. The maximum values occur at different times.

[b] SKI - Swedish Nuclear Power Inspectorate (1991).

[c] SKI - Swedish Nuclear Fuel and Waste Management Co. (1992).

[d] Vieno et al. (1991).

[e] Mobbs et al. (1991) United Kingdom - National Radiation Protection Board (UK-NRPB). Calculated doses from technetium and actinides are not included because solubility limits were ignored. Solubility limits are included in all other safety analyses reviewed here.

TABLE G-3 Relative Doses to Future Individuals From a Repository in Unsaturated Tuff, Loaded with Unreprocessed LWR Spent Fuel

		Relative Dose Rate[a]			
				University of California.[d]	
Radionuclide	Half-Life (years)	Pacific Northwest Lab[b]	Hirschfelder, et al.[c]	$T_W = 10^3$a[e]	$T_W = 10^5$a
^{129}I	1.57×10^{7}	4×10^{-1}	1	4×10^{-1}	8×10^{-1}
^{99}Tc	2.11×10^{5}	1	8×10^{-1}	1	1
^{135}Cs	2.3×10^{6}			2×10^{-1}	3×10^{-7}
^{226}Ra	1.6×10^{3}		4×10^{-12}		
^{234}U	2.45×10^{5}		1×10^{-9}	3×10^{-6}	1×10^{-6}
^{238}U	4.47×10^{9}		6×10^{-7}	1×10^{-6}	1×10^{-6}
^{237}Np	2.14×10^{6}		1×10^{-5}	4×10^{-5}	3×10^{-5}
^{239}Pu	2.41×10^{4}			1×10^{-3}	10^{-40}
^{240}Pu	6.56×10^{3}			2×10^{-4}	$<10^{-100}$
^{242}Pu	3.73×10^{5}		7×10^{-19}	2×10^{-5}	1×10^{-7}
^{243}Am	7.38×10^{3}			3×10^{-5}	$<10^{-100}$

[a] Maximum dose rate from the listed radionuclide divided by the maximum dose rate from the radionuclide that contributes the largest dose. The maximum values occur at different times.

[b] Eslinger et al. (1993). ^{129}I and ^{99}Tc are the principal contributors, with maximum doses about a hundred-fold greater than doses from any other radionuclide.

[c] Hirschfelder et al. (1991, 1992).

[d] Pigford (1990).

[e] T_W is the assumed time for ground water to flow from the buried waste to the environment.

[f] The maximum dose rate from any of these radionuclides is less than 1% of the maximum dose rate from ^{99}Tc.

Similarly, the small penetrations can present a significant impedance to the transport of dissolved radionuclides to the surrounding rock, thereby lowering the calculated individual doses.

Further implications to recycle and transmutation are discussed below.

^{129} Iodine. ^{129}Iodine is the main dose contributor from spent fuel in granite. It is either the most important or second-most important contributor in an unsaturated tuff repository. Iodine is soluble in groundwater. Some iodine will have been released from the UO_2 matrix during reactor operation and will be released to groundwater from defective fuel cladding soon after water enters a defective waste package. In the analyses reported here, the UO_2 matrix is assumed to restructure within a few thousand to a few ten-thousand years, releasing the remaining iodine into groundwater.[2] Iodine forms negative ions in solution and is not expected to be retarded by sorption on the rock. The half-life of ^{129}I is long enough (1.57×10^7 years) that eventually most of it will reach the environment. To reduce the potential individual doses from dissolution and transport in a geologic repository, either the ^{129}I must be transmuted or the reprocessing operations must produce waste forms that release iodine far more slowly than is calculated here for unreprocessed spent fuel. Only the accelerator transmutation of waste (ATW) and the LWR transmutors propose to transmute ^{129}I.

^{99} Technetium. ^{99}Technetium, a fission product, is one of the two (the second one ^{129}I) most important contributors to individual doses in unsaturated tuff. Technetium is released by essentially the same mechanisms as iodine. It is not solubility limited in an oxidizing environment. However, a granite repository is expected to resaturate after sealing, and ferrous iron dissolved in the groundwater is expected to restore a reducing environment that promotes low-solubility forms of technetium that would precipitate in or near the waste packages. Consequently, the dissolution source term for technetium is reduced in granite repositories. The calculations by Sweden and Finland take into account the limited solubility of technetium and predict that technetium doses will be several orders of magnitude below the iodine dose from granitic groundwater.

To reduce the potential individual doses that result from dissolution and transport in a geologic repository, either the ^{99}Tc must be transmuted or the reprocessing operations must produce waste forms that release technetium far more slowly than is calculated here for unreprocessed spent fuel. Only the ATW and the LWR transmutors propose to transmute ^{99}Tc.

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[2] In a repository in unsaturated tuff, the spent-fuel matrix may eventually oxidize if sufficient air or air-saturated groundwater can leak through penetrations in failed containers. In the Swedish analyses for granite repositories, the spent fuel is conservatively assumed to oxidize and release its soluble fission products as a result of alpha radiolysis. In the Canadian repository program, it is expected that alpha radiolysis will not be sufficient to oxidize and restructure the spent uranium fuel, so release of iodine from the fuel matrix is expected to be very slow, limited by the solubility-limited net dissolution of the uranium fuel matrix. Thus, in the Canadian estimates for a granite repository, releases of most of the radionuclides from spent fuel to ground water are expected to be controlled by the slow rate of net dissolution of the unaltered fuel matrix, so the doses predicted for iodine and many other radionuclides are much smaller than predicted in the analyses by Sweden and Finland.

^{135} Cesium. ^{135}Cesium is predicted by Sweden and Finland to be the second-most important contributor in a granite repository. It is also important in the predictions by the United Kingdom's National Radiation Protection Board. Dissolution of cesium is not expected to be limited by cesium solubility. In unsaturated tuff ^{135}Cs is predicted to be an important contributor for pathways that have water travel times to the environment of a few thousand years, third in importance after iodine and technetium. For the longer water travel times (about 10^4 to 10^5 years) predicted from the calculated infiltration rates at the proposed Yucca Mountain repository. Cesium sorption and retardation may delay the transport of cesium to the environment sufficiently for most of the ^{135}Cs to decay in transit. Transmutation of ^{135}Cs would be particularly difficult because of the intense radiation field from ^{137}Cs and because stable ^{133}Cs would absorb neutrons to produce more ^{135}Cs. Separation of the cesium isotopes prior to transmutation would avoid the problem of generating more ^{133}Cs, but isotope separation in the intense radiation field ^{137}Cs would be difficult.

To reduce the potential individual doses from ^{135}Cs in a geologic repository that does not have sufficient water travel time and retardation for ^{135}Cs decay, there appear to be three options: (1) solve the difficult problem of isotope separation and transmutation of ^{135}Cs, (2) produce waste forms that release cesium far more slowly than is calculated here for unreprocessed spent fuel, or (3) store separated cesium for several hundred years until ^{137}Cs has decayed, then proceed with isotope separation and transmutation. Only the ATW considers isotope separation and transmutation of ^{135}Cs as an option to be pursued.

^{231} Protactinium. The data in Tables G-2 and G-3 show that there would be relatively little reduction in the doses to individuals by transmuting only the actinides. However, if the more significant problems of the long-lived soluble fission products are solved, as discussed above, reducing the potential individual doses from dissolution and transport of the actinides would become important. In the calculations for granite repositories, the main actinide contributor is ^{231}Pa, a decay daughter of ^{235}U. It grows in slowly in separated uranium, with a time constant of about 50,000 years. The long-term doses from ^{231}Pa have not been addressed in the calculations for a tuff repository. There is no reason to expect that the importance of ^{231}Pa in tuff would be significantly less than calculated for disposal of spent fuel in granite.

There would not be enough ^{231}Pa in spent fuel to justify its transmutation, but transmutation of its precursors ^{235}U, ^{239}Pu, and ^{243}Am would remove the potential problem of ^{231}Pa. However, transmutation requires reprocessing. Transmutation of the separated TRUs would eliminate ^{239}Pu and ^{243}Am. By recycling the separated uranium to facilities that enrich uranium to fuel LWRs, much of the ^{235}U could eventually be transmuted in the LWRs. This would require continuing use of LWRs while constructing and operating transmutor systems. About 30% of the ^{235}U recycled would appear in the depleted uranium from isotope separation. Long-term build-up of ^{231}Pa in the resulting depleted uranium would continue as a significant environmental issue. In the time scale of a few hundred thousand years and longer, it could present more significant hazards from surface or near-surface dissolution and transport than is expected from the mill tailings from producing new uranium fuel for LWRs.

However, none of the transmutation proposals contemplate recycling the recovered uranium. Instead, the flowsheets proposed by the transmutation proponents to reprocess LWR spent fuel, whether pyrochemical or aqueous, would separate uranium from the other actinides and store it above ground. If that is to be the ultimate fate of the uranium, the long-term individual doses resulting from growth of ^{231}Pa and its ultimate dissolution in surface water could surely be greater than the doses calculated for that same amount of ^{231}Pa that would form in deep geologic disposal in the reference once-through fuel cycle. In this sense, implementing the transmutation proposals that would leave uranium from spent fuel on the earth's surface indefinitely could reduce the overall safety of waste disposal. Even if the separated uranium is recycled to isotope separation, the hazard from the long-term build-up of ^{231}Pa in the resulting depleted uranium, if stored on the surface, may exceed the hazard from ^{231}Pa in spent fuel in a geologic repository. The relative hazards can be and should be calculated.

Other Transuranic Actinides. All of the transmutation proposals reviewed herein would transmute the isotopes of neptunium, plutonium, americium, and curium that appear in LWR spent fuel. The advanced liquid-metal reactor (ALMR) transmutor would transmute only these TRUs; the ATW-1, -2, and -3[3] and the LWR would transmute the TRUs and selected fission products. Transmuting only the TRUs would result in relatively little reduction in the maximum individual radiation dose from dissolution and groundwater transport of waste buried in granite or other reducing rock, because these doses would continue to be dominated by the soluble fission products. Transmuting neptunium could be of benefit to a repository in unsaturated tuff, if the effective solubility of neptunium within failed waste containers is as high as assumed in recent studies (see discussion in section *Long-Term Performance Effects*).

Assuming that transmutation of TRUs could be justified, the most important TRUs whose transmutation could benefit the water-transport pathway in all proposed repositories are the precursors of ^{226}Ra and ^{231}Pa. Transmutation of the ^{238}Pu in plutonium recovered from LWR spent fuel would eliminate the major source of ^{226}Ra. If the uranium separated from LWR spent fuel were buried with the reprocessing waste, the long-term dose from ^{226}Ra[4] would be about seventyfold less than from spent fuel. If the separated uranium were stored on the surface, as proposed in the reprocessing flowsheets, and if the ^{238}Pu were transmuted, the amount of ^{226}Ra that could be formed would be seventyfold less than in LWR spent fuel because ^{238}Pu would not be present. However, if this uranium were not recycled but left on the earth's surface, the lack of geologic isolation could result in a greater long-term dose from ^{226}Ra than calculated for the geologic disposal of spent fuel.

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[3] The ATW-4 transmutor would operate as a self-sustaining thorium breeder. It would not be fueled with TRUs from LWR spent fuel. It would transmute the thorium and all transthorium actinides.

[4] This assumes that uranium is dissolved in groundwater and transports to the environment within a few hundred thousand years, so that the main source of ^{226}Ra would be from decay of the ^{234}U in spent fuel. If the time for uranium transport to the environment is of the order of a million years or more, transmuting the ^{238}Pu would be of little benefit in reducing the dose from ^{226}Ra. The dose from ^{226}Ra reaching the environment would then be governed by the transport of 4.47×10^9-year ^{238}U.

No transmutation projects have proposed transmutation of ^{235}U, the precursor of ^{231}Pa. However, transmutation of ^{235}U in spent fuel, to avoid the build-up and transport of ^{231}Pa, could benefit the long-term performance of geologic repositories.

^{237}Np would be next in importance to dissolution-transport in granite or other reducing rock. It could be more important in unsaturated tuff, as discussed above. Transmutation of neptunium alone would result in only about a fourfold reduction in the long-term dose from ^{237}Np. Decay of untransmuted ^{241}Am would produce additional ^{237}Np that could transport to the environment.

Of the longer-lived plutonium isotopes, transmutation of ^{239}Pu, ^{240} Pu, and ^{242}Pu would be next most important. The predicted doses from these isotopes are of concern only for very short groundwater travel times, such as considered in the Finnish analysis (Table G-2) and in the analysis for 1,000-year water travel time in unsaturated tuff.

^{243}Am and the curium isotopes are not predicted to contribute significantly to doses from TRUs from the dissolution-transport scenarios.

Waste-Form Benefits From Chemical Reprocessing. New waste forms specifically tailored to retain these fission products much longer within the repository offer the potential for improved performance. Benefits could result regardless of transmutation. Candidate waste forms suggested in earlier studies (Pigford et al., 1983) are silver iodide; compounds of separated technetium and separated neptunium, each in thick ferrous containers; and pollucite for cesium. Radiation degradation would need to be investigated. However, the cost of chemical processing solely for waste improvement (i.e., with no recycle of actinides or fission products to reactors) would be staggering. If a U.S. commercial reprocessing plant were to operate with a unit cost of $2,000/kgHM (mostly uranium) in the spent fuel (compare Chapter 7), with no return for recycling uranium and plutonium fuel, the total cost for reprocessing the 63,000 Mg of spent fuel destined for the first U.S. repository would be $126 billion.

Important Radionuclides Contributing To Long-Term Individual Doses From Intrusion Scenarios For The Reference Once-Through Fuel Cycle

Intrusion scenarios, such as inadvertent exploratory drilling, magmatic intrusion from volcanic activity, etc., could bring portions of the buried waste packages to the surface. Also, intrusion by drilling into a repository in unsaturated rock could result in waste cuttings falling through an empty borehole to a lower aquifer, with a correspondingly shorter time of transport of dissolved species to the environment. Here the important radionuclides that could contribute to individual doses are those discussed for the dissolution-transport scenario.

Individual doses to people from waste cuttings brought to the surface could be quite large (Eslinger et al., 1993). If the probabilities of intrusion are low, a risk criterion that considers both dose and probability is appropriate. The radionuclides of highest radioactivity are generally considered to be those of greatest risk. The most important radionuclides for early intrusion, within a few hundred years after emplacement, are ^{90}Sr and ^{137}Cs. The isotopes of plutonium and americium would be important thereafter, until a few hundred thousand years when ^{226}Ra and ^{231}Pa dominate the radiotoxicity of the spent-fuel waste.

Calculated individual doses and risks for assumed intrusion into a granite repository loaded with LWR spent fuel, at 100 years and 10^5 years after emplacement, are listed in Table G-4 (Mobbs et al., 1991). The calculated risk from intrusion is 1.8×10^{-7}/yr for intrusion at 100 years and 2.1×10^{-9}/yr for intrusion at 10^5 years. The same study calculates a maximum risk of 5×10^{-5}/yr from the long-term dissolution of spent-fuel waste in ground water and the long-term transport of dissolved radionuclides to the environment. Thus, Mobbs et al. conclude that dissolution-transport is the more important mechanism to consider in assuring long-term public health and safety from geologic disposal. If so, emphasis in transmutation would be directed to the long-lived, soluble fission products, as discussed above.

The Reference Once-Through Uranium Fuel Cycle: Preliminary Estimates Of Conformance With Technical Containment Limits Of The Epa Standard 40CFR191

Even though the present EPA standard 40CFR191 no longer applies as such to the proposed Yucca Mountain repository, it is useful to examine how a conceptual repository in unsaturated tuff containing unreprocessed spent fuel would perform under the technical criteria used in the standard. Barnard et al. (1992) have estimated the probabilities of releasing radionuclides to the accessible environment by a large variety of scenarios, including gaseous releases, aqueous pathways, human intrusion, and others. For a given release mechanism and probability, the curie releases of each radionuclide is calculated and ratioed to the allowable curie release of each radionuclide, if released alone, as specified by EPA. The sum of these radionuclide ratios for a release scenario of given probability is called the "EPA sum." For each scenario, a complementary cumulative probability distribution function (CCDF) is constructed, consisting of a plot of the plot of CCDF as a function of the EPA ratio. The result is compared with a limiting CCDF specified by EPA.

Results based on a composite-porosity model of flow and transport in unsaturated tuff[5] are shown in Figure G-6. The region that would equal or exceed the EPA limit is shown by the shaded area in the upper right corner. Included are curves for gaseous releases (mainly ^{14}C), releases by dissolution and hydrogeologic transport (the "aqueous" release scenario), releases by human intrusion (i.e., "drilling"), and releases by volcanism. Human intrusion includes three different scenarios, all from exploratory drilling:

1. drilling that intersects one or more waste packages and brings radioactive cuttings to the surface;

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[5] The unsaturated tuff of the proposed Yucca Mountain repository consists of porous fractured rock. In the composite-porosity model, it is assumed that there is local equilibrium of water between the pores and fractures, resulting in a long predicted time for water to move from the zone of waste emplacement down to saturated aquifers several hundred meters below.

TABLE G-4 Individual Doses and Risks From Intrusion Scenarios, Granite Repository, Unreprocessed Spent Fuel[a] [b]

		100 years		100,000 years	
Radionuclide	Half-Life (years)	Dose Rate (mrem/yr)	Risk (yr^{-1})	Dose Rate (mrem/yr)	Risk (yr^{-1})
^{99}Tc	2.11×10^{5}	5.0×10^{-1}	1.5×10^{-14}	1.9×10^{-4}	2.5×10^{-14}
^{129}I	1.57×10^{7}	1.8×10^{-1}	5.4×10^{-16}	3.2×10^{-5}	4.2×10^{-15}
^{135}Cs	2.3×10^{6}	2.4×10^{-1}	7.0×10^{-15}	1.3×10^{-2}	1.7×10^{-12}
^{137}Cs	3.01×10^{1}	1.5×10^{5}	1.8×10^{-7}	-	-
^{234}U	2.45×10^{5}	7.3×10^{1}	2.1×10^{-12}	5.0×10^{-1}	6.5×10^{-11}
^{235}U	7.04×10^{8}	1.8	5.1×10^{-14}	1.4×10^{-1}	1.9×10^{-11}
^{236}U	2.34×10^{7}	1.9×10^{1}	5.4×10^{-13}	2.8×10^{-2}	3.7×10^{-12}
^{238}U	4.47×10^{9}	2.0×10^{1}	5.9×10^{-13}	5.9×10^{-2}	7.7×10^{-12}
^{237}Np	2.14×10^{6}	1.1×10^{2}	3.1×10^{-12}	3.6×10^{-1}	4.7×10^{-11}
^{238}Pu	8.77×10^{1}	3.6×10^{5}	1.8×10^{-7}	5.1×10^{-1}	6.7×10^{-11}
^{239}Pu	2.41×10^{4}	1.0×10^{5}	1.8×10^{-7}	1.2×10^{1}	1.5×10^{-9}
^{240}Pu	6.56×10^{3}	1.5×10^{5}	1.8×10^{-7}	3.1×10^{-2}	4.0×10^{-12}
^{241}Pu	1.44×10^{1}	1.0×10^{6}	1.8×10^{-7}	9.2×10^{-1}	1.2×10^{-10}
^{242}Pu	3.73×10^{5}	5.4×10^{2}	1.6×10^{-11}	8.9×10^{-1}	1.2×10^{-10}
^{241}Am	4.33×10^{2}	2.0×10^{5}	1.6×10^{-7}	1.8×10^{-1}	2.4×10^{-11}
^{243}Am	7.38×10^{3}	6.4×10^{3}	1.9×10^{-10}	3.4×10^{-1}	4.4×10^{-11}
^{244}Cm	1.81×10^{1}	8.7×10^{3}	2.5×10^{-10}	4.7×10^{-4}	$6.\times\times10^{-14}$
^{245}Cm	8.50×10^{3}	4.8×10^{1}	1.4×10^{-12}	7.3×10^{-4}	9.6×10^{-14}
Total		2.1×10^{6}	1.8×10^{-7}	1.6×10^{1}	2.1×10^{-9}

[a] Repository contains 18,000 Mg unreprocessed spent fuel.

[b] Dose-risk conversion factor = 1.65×10^{-4}rem (1.65×10^{-2}/Sv) recommended by ICRP (1977).

SOURCE: Mobbs et al. (1991).

2. drilling that penetrates to a lower aquifer in tuff, with waste cuttings falling through the open drill open to the lower aquifer; and
3. similar drilling but to a deeper lower aquifer in carbonate rock.

The CCDF total for all scenarios is dominated by release of gaseous ^{14}C[6] . Next in importance, for the hydrodynamic model of Figure G-6, is releases by drilling. The principal radionuclides of importance that can be released to the surface and to the lower carbonate aquifer are the isotopes of plutonium and americium. However, for human-intrusion releases to the tuff aquifer the most important radionuclides, as measured by contributions to the EPA ratio, are ^{14}C and ^{237}Np. In the tuff aquifer the groundwater velocity is low enough and sorption high enough that little of the plutonium and americium reach the environment.

Barnard et al. (1992) have also constructed similar CCDF plots for a different model of hydrologic flow in unsaturated tuff, wherein the groundwater is assumed to move rapidly through a few conductive fractures, without local equilibrium with the porosity of the bulk rock, as shown in Figure G-7. Here hydrogeologic transport of dissolved radionuclides is predicted to be a more important contributor to the overall CCDF than human intrusion. Determining which of the flow models best represents unsaturated tuff at Yucca Mountain must await data from further experiments and from site characterization.

The EPA standard 40CFR191, in specifying a generic limit on allowable cumulative releases from a repository, assumes a "generic repository," whereby all radionuclides released to the local environment appear in a hypothetical river. It assumes that a constant future human population of 10 billion people use a certain fraction of that contaminated water for growing food and for potable water. That fraction is assumed to be the same as the fraction of the total surface waters of the northern hemisphere that are used for potable water and irrigation (Pigford, 1981). A calculated collective dose and risk on this basis for the proposed Yucca Mountain repository would be too tenuous to be useful.

The apparent problem of gaseous ^{14}C in dominating the EPA ratio for spent fuel in unsaturated tuff suggests that special attention be given to potential collective doses from gaseous ^{14}C for this type of waste and repository rock. As noted above, individual doses from ^{14}C are predicted to be very low and do not appear to be a problem. None of the proposed transmutation systems are designed to transmute ^{14}C, and such transmutation would be impossible because of its very small neutron cross section. ^{14}C transmutation seems unnecessary because the spent fuel must first be reprocessed to transmute other species. ^{14}C in reprocessing waste, whether from aqueous or pyrochemical reprocessing, is expected to be in a form not easily converted to gas. However, the waste forms expected to contain ^{14}C from reprocessing have not been specified or have not been sufficiently analyzed for ^{14}C retention.

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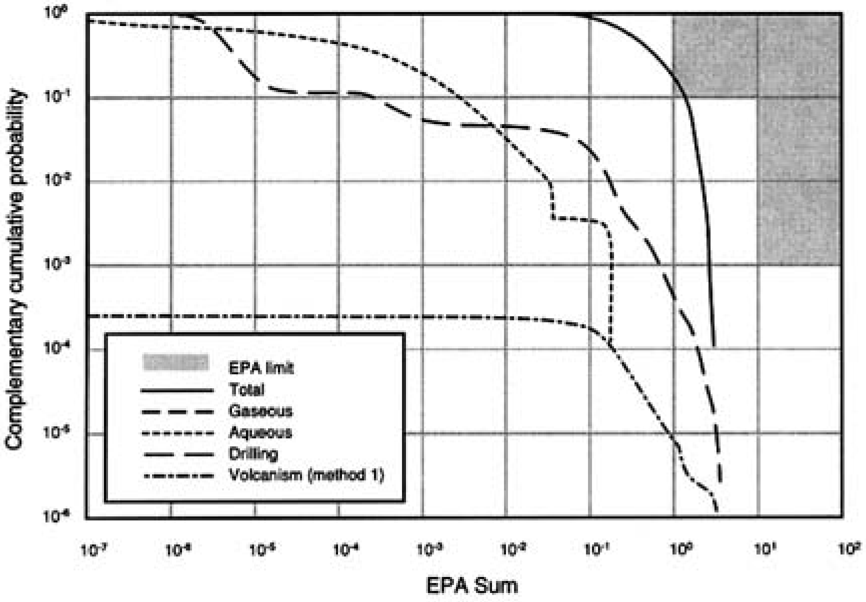
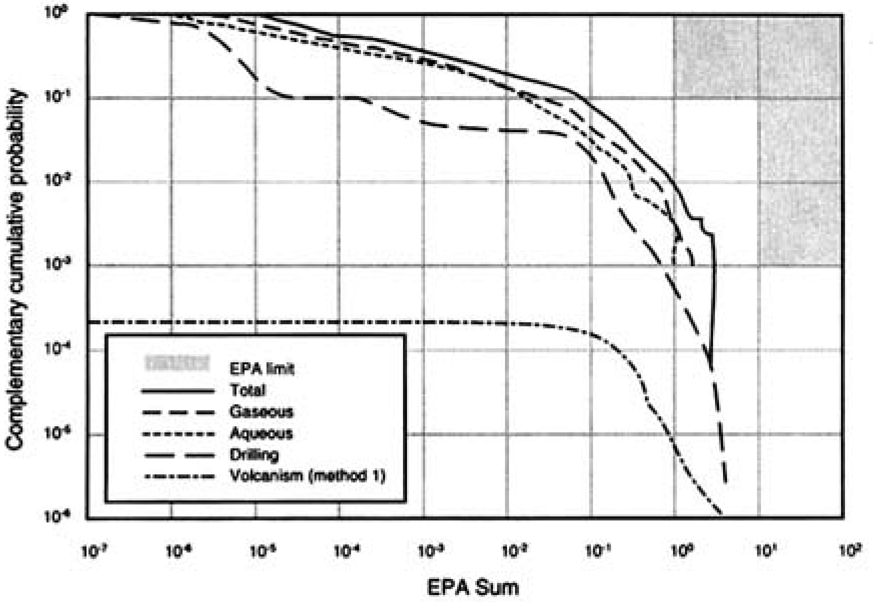
[6] Although there is not a large curie inventory of ^{14}C in spent fuel, essentially all of it is assumed to be released as a gas during a few thousand years. This may be due in part to conservative estimates of the oxidation of the UO_2 in spent fuel to a solid of higher oxidation state, such as U_3O_8.



SOURCE: Barnard et al. (1992)

FIGURE G-6 Overall CCDF for releases assuming the composite model for aqueous transport, spent fuel in unsaturated tuff.



SOURCE: Barnard et al. (1991)

FIGURE G-7 Overall CCDF for releases assuming the conductive-fracture model for aqueous transport, spent fuel in unsaturated tuff.

Individual Doses From Transmutation Fuel Cycles

Benefits from Reprocessing. The simplest transmutation fuel cycle is the reprocessing of LWR spent fuel to separate plutonium and uranium for recycle to LWRs, as is being done in France and planned in other countries. Potential benefits to reduce release of gaseous ^{14}C in an unsaturated-tuff repository, and to reduce collective dose therefrom, are discussed above. Other benefits could arise from reduction in plutonium inventory and from potentially better waste forms,[7] as discussed below.

Benefits from Aqueous Reprocessing and Plutonium Recycle. A reprocessing fuel cycle using aqueous reprocessing could benefit disposal in unsaturated tuff because the borosilicate-glass waste is expected to release soluble fission products more slowly than spent fuel. Also, radioiodine is recovered separately and could be incorporated in a special waste package as AgI. ^{99}Tc in borosilicate-glass waste would be expected to be a dominant radionuclide contributing to individual dose from a repository in unsaturated tuff. Adding technetium separation could result in an improved waste package and lower individual doses from ^{99}Tc. If the conservatively high solubilities of neptunium prevail, as discussed earlier, deploying the neptunium recovery process already developed for aqueous reprocessing could result in a waste form tailored specifically to neptunium, which could mitigate the individual dose from ^{237}Np. Reprocessing to recover and recycle plutonium is predicted to result in over a hundredfold reduction in the risk from plutonium in the human intrusion scenarios, but the total risk from human intrusion is reduced only about twofold because americium and curium are not recycled. The risk from dissolution and migration is still predicted to be the major risk from geologic disposal, about fortyfold greater than the risk from human intrusion. If the waste is to be emplaced in unsaturataed tuff and if the conservative solubility distributions for neptunium, discussed earlier, prevail, adding neptunium separation to aqueous reprocessing could result in a better waste form that would be expected to improve repository performance. Technology for neptunium separation was developed during the 1970s.

Benefits From Reprocessing and Transmutation in ALMRs. A 1991 evaluation of TRU burning in ALMRs by the Electric Power Research Institute (Thompson and Taylor, 1991; Wilems and Danna, 1991) included analysis of the new waste forms that would result from pyroprocessing of spent fuel from LWRs and from TRU-burning ALMRs. Lee and Choi (1991) revised and updated the Institute's analysis of waste forms, to size waste packages at comparable rates of decay-heat generation. Wastes from both aqueous and pyrochemical reprocessing of LWR spent fuel were considered, as well as waste from pyrochemical reprocessing of ALMR spent fuel. They estimated the rates of dissolution of radionuclides from the new waste forms when the radionuclides are contacted with groundwater in a repository in unsaturated tuff. They

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[7] Whether reprocessing yields a better waste form than spent fuel depends much on the geochemical environment of the repository. Reprocessing waste, such as borosilicate glass, may be a better waste form for a repository in unsaturated rock, but spent fuel may be superior in a repository in saturated rock, where a reducing environment is expected.

used release models developed for the Yucca Mountain project and assumed inventories resulting from 0.1% loss of TRUs to waste in each reprocessing cycle. Elemental solubilities developed by the Yucca Mountain project were used, together with solid-solid alteration rates for spent fuel and borosilicate glass waste. For entirely new waste forms, such as the proposed copper-matrix waste from pyrochemical reprocessing, release rates were based in part on oxidation rates reported in the literature. Release rates of soluble species from waste packages would be further attenuated by the assumed slow rate of intrusion of groundwater into a waste container, based on the "wet-drip" water contact mode proposed by the Lawrence Livermore National Laboratory.

From these studies, Lee and Choi (1991) conclude that dissolution rates from reprocessing waste packages are approximately the same as from LWR spent fuel. Even though TRUs have been reduced in inventory by over two orders of magnitude, solubilities rather than inventories control the predicted release rates of low-solubility species from a given waste package.[8] Thus, for the amount of waste that has resulted from a given generation of thermal energy, the predicted doses and risks from dissolution and transport of solubility-limited species to the environment would not be significantly lower for the ALMR wastes that contain lower inventory of TRUs, as compared to spent LWR fuel waste. However, the lower inventories of TRUs in the waste packages would benefit the intrusion scenarios.

Using the dissolution models and parameters developed by Lee and Choi (1991), Hirschfelder et al. (1992) predicted long-term individual doses from using contaminated water from a lower aquifer for a repository in unsaturated tuff. Hydrogeologic transport was based on composite porosity models of groundwater flow, calculated to a distance of 5 km from the emplaced waste. In this analysis, waste from pyrochemical reprocessing was assumed. Maximum doses to future individuals from using contaminated groundwater[9] were calculated, extending to 1 million years. Calculated individual doses for ALMR-transmutation fuel cycles were compared with doses for a reference repository containing LWR spent fuel. It was assumed that temperature limits of the waste package and nearby rock allowed no more than 57 kW/acre of decay heat. Consequently, for the transmutation fuel cycles, the smaller inventory of TRUs and the correspondingly smaller decay heat at the time of emplacement allowed waste from the generation of a larger amount of electrical energy to be emplaced in the same repository area. An additional scenario was calculated assuming that strontium and cesium fission products are separated in reprocessing and stored for a few hundred years. Removing the intense decay-heat source from ^{90}Sr and ^{137}Cs would allow a far greater amount of waste to be emplaced in a given repository area for the same areal heat loading. The stored cesium would eventually have to be emplaced in a geologic repository because of the inventory of long-lived

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[8] Ultimately, if the inventory of a given species in a waste solid is reduced sufficiently, the rate of release from the waste matrix will control its dissolution rate, rather than the rate of dissolution of a precipitate of that species. However, the alteration and dissolution models predict that for 0.001 percent reprocessing loss, the rate of dissolution of uranium, neptunium, plutonium, and americium will be controlled by solubility (Pigford, 1990a).

[9] At the site of the proposed repository at Yucca Mountain, there is no surface water into which groundwater can discharge.

^{135}Cs. The ^{135}Cs is evidently below NRC Class C limits for storage as Class C LLW. However, as discussed earlier, the dissolution and transport of ^{135}Cs can contribute such significant individual doses when emplaced in deep geologic storage. Therefore, it is questionable that storing that amount of ^{135}Cs as LLW could be acceptable if subjected to similar safety analyses.

Relative values of the maximum dose rates for these three scenarios are listed in Table G-5. The maximum dose rates for the reprocessing-transmutation fuel cycle of the ALMR, without strontium-cesium separation, are near or slightly below those for unreprocessed spent fuel, reflecting benefits from improved waste forms. Adding strontium-cesium separation considerably increases the inventory of fission products in the repository and increases the number of waste packages. Even the doses from long-lived TRUs such as ^{242}Pu and ^{237}Np are greater than those for the reference LWR fuel cycle, even though the inventories are lower. Release of plutonium and neptunium are solubility limited. The release of ^{242}Pu and ^{237}Np increase with increased total loading because of the greater number of waste packages, the greater total surface area of waste packages for mass transfer to ground water, and the greater effective volumeric flow rate of ground water into which plutonium and neptunium can dissolve. The largest increases are from the long-lived soluble fission products ^{129}I and ^{99}Tc. The maximum dose rate for ^{129}I in reprocessing waste with strontium-cesium separation is predicted to be about twentyfold greater than for the reference fuel cycle. The relative peak dose from ^{237}Np in unsaturated tuff would increase and could exceed those from ^{129}I if the more conservative solubility data for neptunium, discussed earlier, were used.

These calculations show that there could be a penalty in increased individual dose if transmutation were employed so that waste from a greater amount of electrical energy generation could be loaded in the same emplacement area of the proposed Yucca Mountain repository. Collective dose from these increased loadings would also increase. However, this same increase in collective dose would occur if the increased loading were emplaced instead in a second repository with the same characteristics of the proposed Yucca Mountain repository.

The present approach of the Yucca Mountain Project is to use decay heat to maintain a dry atmosphere at the surfaces of waste containers for thousands of years, with loadings of unreprocessed spent fuel. If that same amount of spent fuel were loaded into a smaller area, to increase the time for a dry repository, individual and collective doses would not necessarily increase. However, if the remaining repository area now designated for Yucca Mountain and the additional area that could be developed were loaded with additional waste produced by power-generating transmutors, individual and collective doses from some radionuclides could increase. Thus, proposals to delay the need for a second repository by implementing transmutation and burying transmutation waste in Yucca Mountain could make it more difficult for the proposed repository to meet a dose or risk limit. However, individual and collective doses would increase if the remaining repository area and the available area were loaded with additional spent fuel, thereby delaying the need for a second repository.

TABLE G-5 Relative Doses to Future Individuals From a Repository in Unsaturated Tuff: Effects of Transmutation, Increased Repository Loading, and Separation of Strontium and Cesium

	Relative Peak Dose Rate[a]		
Radionuclide	Spent Fuel[b]	Waste From Reprocessing and Transmutation[c]	Waste From Reprocessing, Transmutation, and Sr-Cs Separation[d]
^{129}I	1	9×10^{-1}	2×10^{1}
^{99}Tc	7×10^{-1}	4×10^{-1}	7
^{237}Np[e]	1×10^{-5}	2×10^{-6}	3×10^{-5}
^{242}Pu	7×10^{-19}	1×10^{-19}	1×10^{-18}

[a] Maximum dose rate from the listed radionuclide relative to the maximum dose rate from ^{129}I from spent fuel. The individual doses are calculated for dissolution of radionuclides from waste solids and their hydrogeologic transport to the 5-km environment.

[b] Waste from LWR spent fuel, loaded to design limit of decay-heat rate per unit repository area at time of emplacement.

[c] Waste from pyrochemical reprocessing, loaded to the same areal-heat-loading design limit as for spent fuel.

[d] Waste from pyrochemical reprocessing and separation of strontium and cesium for surface storage, loaded to the same areal-heat-loading design limit as for spent fuel.

[e] The relative peak doses from ^{237}Np in unsaturated tuff would increase and could exceed those from ^{129}I if the more conservative solubility data for neptunium, discussed earlier, were used.

SOURCE: Hirschfelder et al. (1991, 1992).

Effect Of Transmutation On Meeting The Technical Containment Limits Of The EPA Standard 40Cfr191

Barnard (private communication, 1993) has calculated the effect of reprocessing and actinide transmutation on the probabilistic distribution of curie releases by aqueous pathways for a conceptual repository in unsaturated tuff. The results are shown in Figure G-8, expressed as the complementary cumulative probability as a function of the EPA sum. Three curves are shown: one for unreprocessed spent fuel, another for waste from aqueous reprocessing of spent fuel and transmutation in ALMRs with pyrochemical reprocessing of ALMR spent fuel, and a third for pyrochemical reprocessing of both LWR and ALMR spent fuel. All curves are based on the same generation of electrical energy. The calculations were made on the basis of the composite-porosity model of hydrogeologic transport in the unsaturated zone.

The EPA aqueous-pathway sums for reprocessing and transmutation are about the same as for unreprocessed LWR spent fuel for probabilities of 0.1 and greater. However, for a probability of 0.001 the EPA sum is about fivefold smaller for reprocessing and transmutation. The calculated differences in EPA sums for aqueous and pyroprocessing of LWR spent fuel are small.

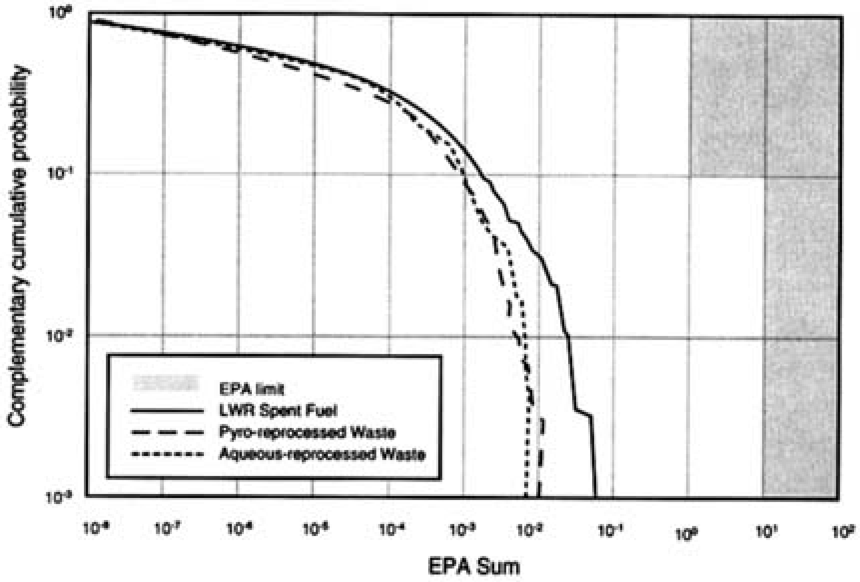
According to these calculations, the greatest benefit from reprocessing LWR spent fuel results from the elimination of gaseous ^{14}C, as may be seen by comparing Figures G-6 and H-8. Although gaseous ^{14}C is emitted to the atmosphere from conventional aqueous reprocessing plants, it is assumed for this discussion that the proposed new high-recovery processes proposed by transmutation proponents, whether aqueous or pyrochemical, would include recovery of ^{14}C into a new waste form of low volatility, suitable for geologic disposal. This ^{14}C benefit results entirely from reprocessing. It would occur whether or not transmutation were also carried out.

Barnard and Lee (1992) calculated the curves of complementary cumulative probability versus the EPA sum for human intrusion, as affected by reprocessing and transmutation, for a conceptual repository in unsaturated tuff. The results for surface releases, based on the composite-porosity model of hydrodynamics in unsaturated tuff and equal electrical energy from reprocessing and nonreprocessing options, are shown in Figure G-9. Three curves are shown: one for unreprocessed spent fuel, another for waste from aqueous reprocessing of spent fuel and transmutation in ALMRs with pyrochemical reprocessing of ALMR spent fuel, and a third for pyrochemical reprocessing of both LWR and ALMR spent fuel. None of the curves would exceed the EPA standard. For probabilities greater than about 10^{-4}, the magnitudes of the releases are lower for repositories containing waste packages from either of the reprocessing options. For probabilities of about 10^{-4} and lower, where the largest predicted releases occur, the predicted releases are about the same for the three scenarios.

Comparing Figures G-8 and G-9, surface releases from human intrusion would be greater, for a given probability, than aqueous-pathway releases for unreprocessed spent fuel, but less for waste from reprocessing and transmutation. For probabilities greater than about 10^{-4}, the EPA sums for surface releases from human intrusion would be lower for transmutation wastes, but the EPA sums would then become dominated by releases from the aqueous pathways. Thus, the reduction in overall EPA sums due to transmutation reduction in actinide inventory is small, less than an order of magnitude. This assumes the composite-porosity hydrodynamic model of the unsaturated zone. For the more rapid hydrodynamic transport in conductive fractures, without local equilibrium with the rock matrix (see Figure G-7), releases via the aqueous pathways are predicted to be more important than those from human intrusion, and separation and transmutation would not be expected to result in as large a reduction in releases as for the composite-porosity model. However, in either of the predictions using the two hydrodynamic flow models, a significant reduction in calculated EPA ratios is predicted due to elimination of gaseous ^{14}C. This is a consequence of the improved waste forms for ^{14}C, a result only of fuel reprocessing.

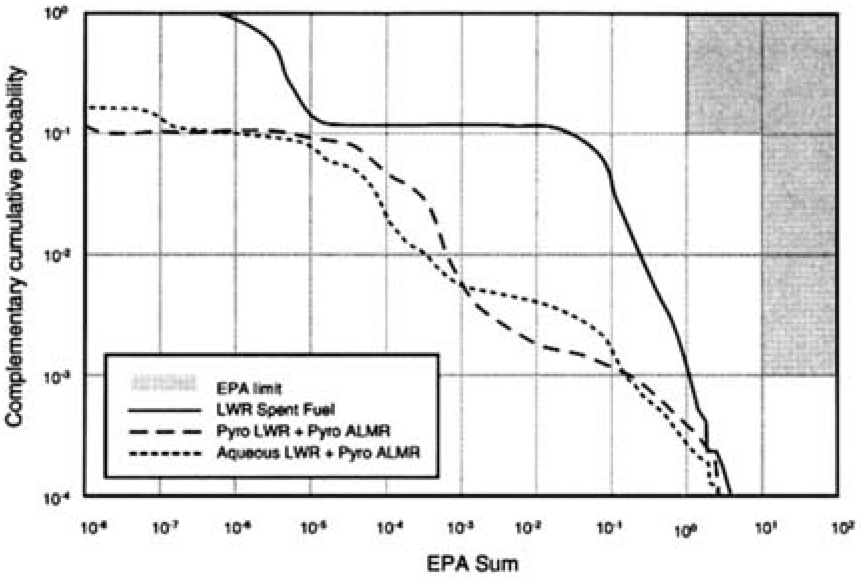
Additional reductions in EPA sums would occur if soluble ^{99}Tc and ^{129}I were also transmuted.

In summary, the calculations by Barnard and Lee (1992) for composite-porosity hydrodynamics show that aqueous-pathway curie releases, as measured by the EPA sums, approach the EPA limit most closely for a probability of 0.001, where aqueous-pathway EPA sums are predicted to about fivefold smaller for transmutation waste. A larger reduction would



SOURCE: Barnard and Lee (1992)

FIGURE G-8 Comparison of aqueous releases for LWR spent fuel and S&T waste in unsaturated tuff.



Source: Barnard and Lee (1992).

FIGURE G-9 Comparison of human-intrusion surface releases for S&T waste and LWR spent fuel in unsaturated tuff (equal generation of electrical energy).

arise only from reprocessing, which could result in a better waste form for retention of ^{14}C that could otherwise be a gaseous release from spent-fuel waste.

Summary And Conclusions

Implementation of S&T would have two direct effects on the repository:

- substantial reduction of the amounts of certain radionuclides in the repository wastes, with the identity of the radionuclides and degree of removal being dependent on the spent-fuel reprocessing technology employed; and
- alteration of the waste form from spent nuclear fuel (rods of actinides and fission products encased in metal) to a monolithic HLW form designed for repository conditions and contained in an unirradiated metal canister including waste forms tailored for containment of specific spent-fuel constituents (e.g., ^{14}C).

These direct effects have diverse impacts on the postclosure behavior of the repository. While these impacts are not yet fully elucidated, they are well enough understood to support the following conclusions:

- *Implementation of S&T would significantly affect the thermal attributes of waste emplaced in a repository*. Removal of actinides only would reduce near-term thermal power by about 20% and reduce the total heat released to the geology surrounding the repository over the long-term by a factor of 4.
- *The removal of the actinides from the material being emplaced in the repository allows 4 to 5 times more waste to be emplaced in a given area of the repository*. While increases in emplacement density can be achieved by other engineering measures, the increase resulting from S&T is additive to these other measures. The increase in emplacement efficiency could extend the life of the repository by four-to fivefold, thus deferring the need to undertake contentious and expensive activities associated with a second repository until about the twenty-second century. In addition, it offers a mechanism to lower the temperature of the repository rapidly (relative to spent fuel) if a "cool" repository concept should be deemed desirable. However, it would also eliminate the ability to establish a "hot" repository that might be employed to keep a repository constructed in unsaturated rock dry for an extended time, as has been proposed for Yucca Mountain.
- *Removal of actinides and strontium and cesium may increase the capacity of the repository by factors ranging from 10 to 40*. This would greatly postpone the need to site, license, and pay for another repository. While operations are simplified, the repository would have to remain open over extremely long time periods. Most important, a MRS-license facility must be sited, built, and operated to store strontium and cesium for hundreds of years.
- *The incremental advantages of removing strontium and cesium in addition to the actinides (i.e., no thermal pulse, very large capacity) will probably not justify the incremental disadvantages (i.e., the need for a very-long term MRS-like facility, the need to separate and*

process strontium and cesium waste), although additional quantification of benefits and penalties is required to substantiate this.

- *The impact of S&T on the long-term risk from the repository is heavily dependent on (1) the specific repository being considered, (2) the types of radionuclides that are removed during reprocessing, and (3) the release scenario being considered*. A generalized summary of the effects of S&T on long-term repository risk is given in Table G-6. It should be noted that the contents of Table G-6 are somewhat dependent on the repository host medium being considered (dry versus wet, oxidizing versus reducing) and the measure of risk used (individual versus population).
- *It is to be emphasized that a S&T scenario in which the need for a repository is eliminated is considered to be highly unlikely if not absolutely impossible*. All current approaches to S&T result in a waste containing significant amounts of radionuclides that are extremely difficult to separate (i.e., require isotopic separation) or that are not amenable to transmutation. Elimination of a repository would require an extremely diverse and sophisticated combination of chemical and isotopic separation technologies in concert with both transmutation and alternative radionuclide disposal technologies such as extraterrestrial disposal.
- *The benefits of S&T (which is defined as enhanced reprocessing to recover essentially all radionuclides that would otherwise report to repository wastes) to long-term repository risk can largely, if not totally, be achieved by employing basic reprocessing (i.e., recovery of about 99% of the uranium and plutonium in the spent fuel)*. This is because the major benefits are reducing heat generation and reducing the toxic actinide content (which can be largely achieved by removing most of the plutonium) and improving the waste form for the residual (which results from reprocessing, irrespective of actinide decontamination levels).

The committee recommendations pursuant to the above conclusions are as follows:

- The Department of Energy should consider the removal of actinides as one option in its broader systemic evaluation of the thermal strategy for Yucca Mountain.
- Pursuit of a HLW repository should be continued.
- The benefits of S&T should continue to be studied as part of the continuing evaluation of repository performance. This should include explicit consideration of the optimum recovery of various radionuclides.
- S&T technology should continue to be developed in an orderly manner, and by the turn of the century it should be brought to the point where preferred technologies could be selected and demonstration projects initiated if deemed appropriate. This development should be closely coordinated with continued development of the ALMR and its attendant nuclear fuel cycle.
- The design of the repository should incorporate features that would allow spent fuel to be readily retrieved and reprocessed and the resulting HLW to be emplaced at a higher effective density.

TABLE G-6 Summary of the Effect of Partitioning-Transmutation on Repository Risk

	Release Mechanism		
Waste Component Released	Dissolution and Migration	Human Intrusion	Gaseous Release
Actinides[a]	Small reduction in expected risk; Substantial reduction of already small risk from low-probability, high-consequence release scenarios; Significant reduction in individual dose	Reduction of risk approximately proportional to reduction in actinide concentration	No significant effect
Iodine and Technetium	Significant reduction in an already-small risk	Small reduction in expected risk	No significant effect
Carbon-14	No significant effect	No significant effect	Major reduction in potentially limiting species that poses a very small individual risk
All Species	High-level waste form is expected to be more resistant to degradation in the repository than spent fuel, thus reducing radionuclide release rates and consequential risk. The magnitude of the benefit depends on the specific radionuclide, with more soluble species being benefitted more.		

[a] The relative peak doses from ^{237}Np in unsaturated tuff would increase and could exceed those from ^{129}I if the more conservative solubility data for neptunium, discussed earlier, were used.

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Appendix H

Nuclear Proliferation Issues

Introduction

Scope and Organization

The purpose of this appendix is to provide background information and supplementary detail to support the evaluation in Chapter 6, *Proliferation Issues* section, concerning nuclear proliferation issues raised by the possible deployment of a separations and transmutation (S&T) system. That evaluation uses the light-water reactor (LWR) once-through fuel cycle as the baseline, taking into account the U.S. policies for nonproliferation, the disposition of plutonium, and nuclear energy. This section provides a brief introduction to the nonproliferation policy context and summarizes how the LWR once-through cycle fits into that context, in particular, the relation to the safeguards system of the International Atomic Energy Agency (IAEA).

The following section, *Nonproliferation Policy and International Safeguards*, reviews the IAEA safeguards system and the U.S. nonproliferation policies that affect the use of S&T technologies and facilities. The review emphasizes the period since 1970 and highlights major nonproliferation successes and failures that had an impact on U.S. policies for nonproliferation and energy. At present, the credibility of the IAEA safeguards system and of the agency itself are being severely tested by two situations: (1) the discovery in 1991 of a covert nuclear weapons program in Iraq as a result of the special inspections conducted there by the IAEA from 1991 to the present under U.N. Security Council mandate and (2) the current difficulties in achieving a satisfactory safeguards implementation with North Korea, amid concerns that it may have a clandestine nuclear weapon effort. The section discusses the impact of the two situations on nonproliferation policy and reviews suggestions for strengthening IAEA safeguards.

The final section of the appendix, *Assessment of Proliferation Issues for S&T Systems*, provides detail on the nuclear proliferation issues that are raised by S&T systems, emphasizing the ways in which an S&T deployment could either increase or minimize the risk of nuclear weapon proliferation compared with the once-through LWR fuel cycle, assuming both are implemented under safeguards. The section treats reprocessing, enrichment, and reactor facilities and technologies generically and also assesses four specific S&T proposals.

The LWR Once-Through Fuel Cycle as the Nonproliferation Baseline

The general objective of U.S. nonproliferation policy is to prevent the misuse of civil nuclear facilities and materials for military purposes and to discourage dedicated nuclear weapons programs.[1] This and more specific objectives are implemented through a web of formal and informal arrangements—treaties, agreements, laws, voluntary guidelines, understandings, and working relationships—that have grown over half a century and involve a variety of multinational and international institutions, governmental organizations, and private groups.

The key international organization for nonproliferation is the IAEA, which was chartered at the United Nations in 1957 in the wake of the U.S. "Atoms for Peace" initiative. The agency has dual responsibilities to promote the peaceful uses of nuclear energy and to guard against the diversion of the products of its use for military purposes.[2] As discussed in the next section, the IAEA instituted a system of safeguards that feature inspections of civil nuclear facilities in IAEA member nations.

Efforts to limit the spread of nuclear weapons were greatly strengthened in 1970 with the establishment of the Nuclear Nonproliferation Treaty (NPT).[3] Over 140 nations are parties to the NPT as nonnuclear-weapon nations, which agree not to acquire nuclear weapons or the means to produce them and to place all nuclear facilities and stores of fissionable materials under IAEA safeguards. Parties as nuclear weapon nations agree to share the technology for peaceful uses of nuclear energy and to refrain from helping nonnuclear-weapon nations to acquire nuclear weapons.

Nonproliferation efforts have focused on controlling fissionable weapon materials—either plutonium or highly enriched uranium (HEU). Obtaining such materials in sufficient quantity and purity is recognized as the most challenging task in the development of a nuclear weapon. The task has three types of requirements: (1) *technical*—attaining the knowledge and know-how to enrich uranium or operate a reactor and reprocess the spent fuel to recover the plutonium; (2) *practical* —acquiring trained manpower and equipment to build and operate the medium-to large-scale facilities needed to produce fissionable weapon materials in sufficient quantity and purity; and (3) *political*—controlling the national political and military apparatus and maintaining secrecy or suitable international relations long enough to obtain the required materials and assemble (and perhaps test) the explosive devices. In effect, the three types of requirements, in addition to economic costs, pose barriers to nuclear weapon proliferation.

The LWR proved to be an efficient first-generation nuclear system for electrical power production. Moreover, the fresh LWR fuel contains low enriched uranium (LEU) that presents

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[1] The nonproliferation objectives of many nations would also emphasize that nuclear weapon nations pursue negotiations on effective measures to end the nuclear arms race and reduce their nuclear arsenals. Such objectives are stated in Article VI of the Nuclear Nonproliferation Treaty as an obligation of the nuclear weapon nations.

[2] For a historical review of the IAEA and an extensive discussion of safeguards, see Scheinman (1987).

[3] For a concise and incisive evaluation of the treaty and the history leading up to its signing, see Bunn (1968). See also Willrich (1968). An evaluation of the impact of the NPT on safeguards is provided in Scheinman (1987). Background information on the NPT and a text of the treaty is given in Fischer and Szasz (1985). A recent critique, including issues raised by the covert nuclear weapon in Iraq, is contained in Bunn (1992) and Scheinman (1992b).

a high barrier to nuclear proliferation. The fresh fuel contains only 3-4% of the fissile isotope ^{235}U diluted with nonfissile^{238}U, which is a much lower enrichment than the HEU used for nuclear weapons, and which is generally greater than 90% ^{235}U. The dominant technologies for isotopic enrichment, in particular the gaseous diffusion and gas centrifuge methods, require specialized materials and other high technologies that are not generally available (see *Assessment of Proliferation Issues for S&T Systems* below). A few developing nations—Argentina, Brazil, Pakistan, and South Africa—finally acquired the knowledge and know-how for uranium enrichment during the 1970s (see the following section). However, constructing and operating the large, complex enrichment plants poses a practical barrier to producing HEU, a barrier that was overcome by the five declared nuclear weapon nations but by no other nations until the 1980s.

The plutonium in the LWR spent fuel presents a dilemma. While ^{239}Pu is the best isotope by far for a weapon, all the plutonium isotopes are fissionable by fast neutrons in a nuclear weapon (Mark, 1990) or a "fast" reactor having a high energy neutron spectrum (see Chapter 4). Isotopic dilution, an effective strategy with uranium, is not an effective means to prevent plutonium from being used as nuclear weapon material. The technical proliferation barrier is not nearly so high for plutonium as for ^{235}U, because only chemical separation is needed. However, ^{239}Pu is also an excellent nuclear fuel in reactors. In the 1960s, the major nuclear supplier nations were planning to reprocess their civil-reactor spent fuel to separate the plutonium for recycle in civil power reactors (see Chapter 4). These nations demonstrated plutonium recycle to their LWRs, but anticipated plutonium recycle to fast breeder reactors when such reactors became commercialized in the future.

Normally, the spent fuel rods would be stored in water pools for several years to cool thermally and radioactively. This intense radioactivity presents a formidable hazard but does not prevent chemical separation. The technical knowledge for such separation became generally available during the 1950s. Yet the practical proliferation barrier for medium-to large-scale plutonium reprocessing was (and still is) substantial (see Chapter 3). When the LWR was being introduced commercially, spent-fuel reprocessing was confined to the nuclear weapon nations. In the late 1950s and the 1960s, however, many nations began small research and development programs on reprocessing technology, ostensibly to explore the future use of the recovered plutonium in their civil power reactors. Several of these programs proved to be proliferation seeds, as discussed in the next section.

To minimize the proliferation risk posed by the LWR spent fuel, the fuel might be disposed of permanently in a once-through cycle—isolation of high-level waste (HLW) in geologic repositories had been proposed in 1957. Yet, this would preclude plans at that time for the future civil use of the fissionable resources contained in the spent fuel. As an alternative, the spent fuel could remain in storage pools for an extended period and the disposition (reprocessing or disposal) deferred, provided nations could be assured that the spent fuel would not be diverted and used to obtain plutonium for nuclear weapons. That concern was met successfully by the development of IAEA safeguards, which act as a political/technical barrier, as discussed in the next section.

It is useful to compare the LWR with two other contenders for a first-generation nuclear power standard—namely a heavy-water reactor (HWR) and a liquid-cooled graphite-moderated reactor (LGR). Both have better neutron moderators than a LWR in terms of neutron economy; in fact, a HWR can be operated on natural uranium. However, both can be refueled on-line (i.e., replacing fuel rods while the reactor is still operating; see *Assessment of Proliferation Issues for S&T Systems* below). This makes them attractive for producing weapons-grade plutonium; indeed, HWRs and LGRs were used for that purpose by the nuclear weapon nations. In comparison, the characteristics of a LWR require refueling in a batch mode (i.e., shut-down, typically for an annual change of a third of the reactor core).

A commercial HWR for electrical power production, the CANDU, was developed by Canada and exported to several nations, including India and Argentina. However, its excellent properties for plutonium production raised serious questions about the wisdom of HWR export. Indeed, India used its CANDU to make plutonium for a nuclear explosive device (see the following section). Following that incident, Canada worked with the IAEA to develop a system of safeguards for the CANDU (see the last section of this appendix) which are required by Canada as a condition of sale. The batch refueling characteristics of the LWR, on the other hand, accommodated well to a control regime including the IAEA safeguards being established during the early to mid-1960s. This opened the way for LWR exports to many nations for electrical power production, while maintaining high nuclear proliferation barriers. It was not until the mid-1970s, however, that the LWR once-through cycle with geologic isolation of the spent fuel was adopted by the United States and many other nations.

Nonproliferation Policy And International Safeguards

Overview of Safeguards Concepts

This subsection provides an overview of IAEA safeguards concepts that would apply to a S&T system. Although the IAEA predates the NPT by some 13 years, the treaty had a profound effect on the agency and its safeguards system. The following subsection discusses the comprehensive safeguards basis for NPT nonnuclear-weapon nations as defined in the IAEA Information Circular 153 (INFCIRC 153). It is compared with the varied and much less comprehensive safeguards basis under Information Circular 66, which was developed in the 1960s and applies to IAEA member nations that are not yet NPT signatories.

The objective of the IAEA safeguards system is "… the timely detection of diversion of significant quantities of nuclear material from peaceful nuclear activities to the manufacture of nuclear weapons … and deterrence of such diversion by the risk of timely detection."[4] A related political objective is to provide assurance to the international community that nations are complying with their nonproliferation agreements and that any significant diversion would be detected in a timely manner. The nations enter into safeguards agreements voluntarily; the

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[4] See Fischer and Szasz (1985) for a critique of safeguards and the inspection process by two former IAEA officials. That reference also provides the text of key background documents on safeguards and the NPT.

IAEA has no authority to apply safeguards unless a nation so requests, as NPT signatories are obligated to do.

To meet the objective, the IAEA safeguards system emphasizes *materials accountancy* augmented by *containment and surveillance* as the key verification measures. The materials accountancy concept, historically the primary emphasis of IAEA safeguards, focuses on the quantification of any *material unaccounted for*. Materials accountancy uses random sampling techniques and special technologies (e.g., secure identification tags and seals, instruments to assay nuclear materials). These are implemented with inspections at regular and random intervals, together with extensive maintenance and checking of records to compare expected quantities of nuclear materials with measured quantities (Fischer and Szasz, 1985).

Containment and surveillance technologies and techniques have historically been used by the IAEA to augment the primary materials-accountancy practices, especially where conversion time might be short or material quantities are large. Recently, with the advent of more advanced surveillance equipment and secure communications, such techniques are being considered for a more prominent safeguards role in applications with large facilities for enrichment or for reprocessing and refabricating nuclear fuels, as would occur in S&T applications (Scheinman, 1992a).

The frequency of inspections and the design of random sampling are geared to the quantity and form of the nuclear material under safeguards and to country-specific factors, particularly to the estimated time for a nation to obtain sufficient quantity in such a form that the manufacture of a nuclear explosive device cannot be excluded. For example, spent fuel rods containing plutonium or HEU pose a qualitatively different safeguards problem than the same quantities of nuclear materials in separated bulk form. Using statistical methods, the IAEA aims at a 90-95% probability level for detection of a diversion, with a false alarm rate of less than 5%.

The concepts of timely detection and deterrence by the risk of early detection are central to understanding the system and evaluating its effectiveness (see Scheinman, 1987:165–166). The expected result of applying IAEA safeguards is to confirm that no diversion has taken place. Were the IAEA to discover a possible diversion, the agency cannot by itself take direct corrective action but, rather, is obligated to report the situation immediately to its board of governors and, thus, to the international community. The IAEA member nations would take the appropriate action. Therefore, the likelihood of detection must be certain enough, and the time scale for detection and report of any suspected diversion short enough, that a high risk of early detection affords deterrence.

The goals and criteria for materials accountancy are geared to the detection of quantities of safeguard significance of special nuclear materials for building a single nuclear explosive device, which the IAEA takes to be 8 kg for plutonium and, for uranium of greater than 20% enrichment, an amount containing 25 kg of ^{235}U. These amounts reflect an estimate of how much is likely to be required for a straightforward device design that might be produced by a nation. The publicly available literature on nuclear weapons suggests significantly lower mass requirements for both materials.[5] The mass estimates per nuclear device of 5 kg of plutonium and 15 kg of HEU are used in the next section to assess the proliferation issues raised by various types of nuclear facilities.

Safeguards Under the NPT

Article III of the NPT specifies the key safeguards and inspection provisions of the treaty, which apply to *all* nuclear materials and facilities used for peaceful purposes in nonnuclear-weapon nations, and which all nonnuclear-weapon signatories are obliged to accept. The broad nature of Article III and the experience with pre-NPT safeguards under Information Circular 66 stimulated demands by several nations with advanced nuclear capabilities, including Germany and Japan, that IAEA safeguards be reviewed and strengthened. The resulting IAEA safeguards basis was codified under Information Circular 153 and the related subsidiary agreements and facilities agreements.[6]

The Information Circular 153 implementation is better defined and more uniformly applied than in earlier bilateral safeguards agreements under Information Circular 66 that are still in effect with non-NPT nations, such as India, Israel, and Pakistan. In particular, the safeguards agreements under Information Circular 153 have strengthened and unified provisions for recordkeeping and reporting requirements and for implementing materials accountancy and containment and surveillance technologies and practices. In contrast, safeguards agreements under Information Circular 66 apply only to certain facilities agreed on by the nation and the IAEA and sometimes are limited to nuclear materials that are imported.

Another key improvement of NPT-type safeguards was the adoption of the concept of *diversion for purposes unknown*, which put the burden of responsibility on the nation in question (rather than on the IAEA) to prove that any unaccounted material of significance has not been diverted to military purposes. Inspections were codified to include random as well as regularly scheduled inspections at facilities declared by a nation to contain stores of special nuclear materials or to be capable of their production. Under NPT, Article II, nonnuclear-weapon

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[5] For an implosion weapon with a "tamper" of uranium that retards disassembly, the literature reports the critical mass of a shell of metallic plutonium as 4.5 to 5 kg with a good neutron reflector, versus 8.6 kg for a plutonium shell with no reflector. The critical mass for a shell of metallic 94 percent ^{235}U is reported as 13 to 15 kg with a good neutron reflector versus 26 kg with only a tamper (see Hebel et al., 1978; Paxton, 1964; Paxton et al., 1964). An estimate of 15 kg of HEU is also reported in Alison et al. (1993:118). Some 25 to 30 kg may be needed for a uranium gun-type device. In the secondary stage of a two-stage weapon, 15 to 25 kg of HEU is estimated for devices with a yield of a few hundred kilotons, with more HEU for large secondaries in the multimegaton range (see Cochran et al., 1984).

[6] *The subsidiary agreements* define the details of the safeguards obligations, for example, the frequency of inspection, the information to be shared with the agency, and the recordkeeping required. The *facilities agreements* address key issues such as the material balance areas, the measurement points for safeguards application, the measures that are permitted for surveillance and containment, and related details (see Fischer and Szasz, 1985).

signatories are obliged to declare all such facilities on their territory. IAEA also has authority to make "special inspections" of suspected undeclared facilities, activities, and nuclear materials under Information Circular 153 and, in principle, under the charter, within any IAEA member nation (Blix, 1992; Bunn, 1992; Scheinman, 1992a). Before the 1991 inspections in Iraq, however, neither the NPT nor bilateral safeguards agreements were used by the IAEA to authorize special inspections.[7] The nations did not exchange the intelligence information with the IAEA that might have lead to requests for such inspections.

The NPT, with IAEA safeguards, is the cornerstone of the international nonproliferation effort, which has enjoyed remarkable success. NPT members Germany, Japan, and South Korea are notable examples of nonnuclear-weapon nations with large civil nuclear programs. The NPT nations include most states of Eastern and Western Europe, several of the latter with nuclear programs covered by Euratom safeguards. Moreover, numerous developing nations decided that the NPT would provide a suitable political framework for their small nuclear power programs. However, several nations with sizable nuclear programs remain outside the NPT framework, although all of them agreed to IAEA safeguards for some of their facilities as a price for the ability to import nuclear technology and systems from the major nuclear suppliers. Such facilities that have not been included in the IAEA safeguards regime have been the basis for several nations to attain de facto but undeclared nuclear weapon capability, as discussed below.

Creeping Proliferation and the Effect on Nuclear Policies

The major nuclear nations differ at present on reprocessing and the use of plutonium in civil reactors. In the 1960s, as noted earlier, the United States and a few other nations engaged in the limited reprocessing of LWR spent fuel and the recycle of the recovered plutonium to commercial LWRs and other thermal reactors, looking ahead to breeder reactor deployment. However, many nations were forced to reexamine their policies and plans when India detonated a "peaceful nuclear explosion" in 1974. The event became all the more vexing for Canada and the United States when the public learned that India produced the plutonium for the device using a Canadian-built reactor and fuel of U.S. origin, which had been sold to India without IAEA safeguards (see Spector, 1988).

In the mid-1970s, the United States cancelled its breeder deployment plans and discontinued reprocessing and thermal recycle of plutonium, for both economic and nonproliferation reasons.[8] The United States adopted the LWR once-through fuel cycle with geologic disposal of the spent fuel and encouraged other nations to do the same. Moreover, Congress passed the Nuclear Nonproliferation Treaty of 1978, which restricted the reprocessing of nuclear fuel of U.S. origin. Congress also enacted the Glenn-Symington Amendment to the

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[7] In 1992, the IAEA board of governors reaffirmed the right of inspections under Information Circular 153 at undeclared sites in nonnuclear-weapon nations.

[8] For a penetrating review of U.S. nonproliferation and energy policies up to the mid-1970s, see Farley (1976), Willrich et al. (1973), and Greenwood et al. (1977).

Foreign Assistance Act of 1979, which provided for the cutoff of economic assistance to any nation considered by the Carter administration to be importing a capability for uranium enrichment.

From 1976 to 1979, the nuclear nations, large and small, took part in an International Fuel Cycle Evaluation (INFCE), which reexamined all aspects of the nuclear fuel cycle. The Foreign Assistance Act of 1979, which provided for the cutoff of economic assistance to any nation considered by the Carter administration to be importing a capability for uranium enrichment participants agreed that the once-through fuel cycle with geologic disposal of LWR spent fuel could be a safe practice and that the time scale, if not the desirability, of a cost-effective breeder economy was very uncertain (INFCE, 1980). The European nuclear suppliers suspended their plans to export reprocessing facilities and technology to several developing states. However, France, the United Kingdom, the former Soviet Union, and Japan continued their policies to reprocess civil-reactor spent fuel and store the separated plutonium not needed for breeder reactor research and development.

By the early 1980s, several factors combined to take the heat out of the debate about reprocessing versus once-through for LWR spent fuel: (1) the discoveries of large uranium ore reserves in Australia and Canada; (2) reduced uranium ore requirements due to less deployment of LWRs in the United States and several other nations than had been projected earlier; (3) high costs and continuing technical difficulties of breeder reactors; and (4) agreement that the once-through fuel cycle could, in principle, resolve the issues attending the steadily growing amount of spent fuel in pool storage around the world. In 1982, the Reagan administration relaxed the ban against reprocessing in the United States. However, the change had little practical effect, since reprocessing of LWR spent fuel for recycle was uneconomic for the reasons cited above. Moreover, the restrictions in the 1978 Nuclear Nonproliferation Treaty still applied.

Stability proved to be elusive as a series of public exposures during the 1980s revealed that three additional non-NPT nations—Israel, Pakistan, and South Africa—had attained de facto but undeclared nuclear weapon capability (Spector, 1988). Like India, the three nations used nuclear facilities not under IAEA safeguards and drew heavily on their civil nuclear programs to attain such capability (Spector and Smith, 1990). Also, two other nations, Argentina and Brazil, disclosed their ability to produce enriched uranium. The particulars of the above are summarized as follows:

- In the case of India, the facilities and trained manpower from the nation's civil nuclear program, including a purchased CANDU reactor, played a direct part in the plutonium production for the nuclear device test in 1974 (Spector, 1988:80-119). India subsequently constructed several large reactors and a large reprocessing complex that operate outside IAEA safeguards and is reported to have material for several hundred nuclear weapons (Albright and Zamora, 1989).
- Israel conducted plutonium reprocessing at Dimona from the late 1960s and reportedly has accumulated material for a hundred or more nuclear weapons (Spector, 1988:164-193). The publicly available information from a 1986 expose in the *Sunday Times of London* includes cut-away pictures of sophisticated nuclear devices.
- Pakistan began uranium enrichment in the mid-1980s using the centrifuge method reportedly based on stolen European design information. The facility at Kahuta may have

produced a material stockpile sufficient for tens of nuclear weapons (Albright and Zamora, 1989). In October 1990, the Bush administration was no longer able to certify to Congress that Pakistan did not possess nuclear weapons, which resulted in a cutoff of economic and military assistance to Pakistan under the 1985 Pressler Amendment to the Foreign Assistance Act.

- South Africa indigenously developed the capability for uranium enrichment (Spector, 1988). In 1990, South Africa disclosed that it had constructed six gun-type uranium nuclear weapons in the 1980s, which were destroyed when the nuclear weapon program was abandoned (Albright, 1993b; de Villiers et al., 1993). South Africa recently joined the NPT, accepting full-scope IAEA safeguards.
- Argentina and Brazil, which are not NPT members, also developed the technology and facilities capable of "dual-purpose" uranium enrichment—Argentina using gaseous diffusion and Brazil using the centrifuge approach (Albright, 1989, 1990; Donnelly, 1990). In the late 1980s, however, the two nations ended their long-standing nuclear rivalry and entered into a stringent bilateral nuclear safeguards regime backed up by full-scope IAEA safeguards that have provisions for regular, random, and special inspections (Albright, 1990; Redick, 1990; Krasno, 1992).

Yet, because of the growing number of NPT signatories, this series of exposures did not seriously impugn the credibility of the IAEA safeguard system.[9] Indeed, Argentina and Brazil, and to a lesser extent South Africa, could be regarded as nonproliferation success stories. Moreover, the progressive improvement in safeguards technologies, developed through cooperative programs between the IAEA and various nuclear powers and gradually deployed in nonnuclear-weapon nations, further encouraged the perception that safeguards in NPT nations were adequate. By the mid-1980s, however, U.S. nonproliferation policy began to emphasize coping with rogue nations concomitantly, with less emphasis on the earlier priority of limiting the spread of reprocessing and enrichment capability beyond the European nuclear suppliers, the former Soviet Union, and Japan. The shift of emphasis was accentuated by the recent events in Iraq and North Korea, discussed below.

The Crisis in Safeguards Credibility

The Covert Iraqi Nuclear Weapons Program

After the Persian Gulf War, the IAEA conducted a series of special inspections of facilities and records in Iraq, a NPT signatory since the 1970s. Acting under a U.N. Security Council mandate and benefiting from intelligence information, the IAEA inspection team discovered a covert effort to develop nuclear weapons—in violation of Iraqi treaty obligations (Fainberg, 1993; Davis and Kay, 1992; Thorne, 1992; Donohue and Zeisler, 1992; UN/IAEA,

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[9] There has arisen, however, a growing number of skeptics about IAEA and its safeguards system. See, for example, Bolt (1988) and Leventhal (1987, 1992).

1991). The effort centered on a uranium enrichment capability that was based primarily on the inefficient but technologically straightforward method of electromagnetic isotope separation. The extensive facilities near Al-Tarmiya and Ash-Sharkat, under construction but not yet in operation, were undetected in regular IAEA inspections prior to the war (see Pilat, 1992).

Iraq was perhaps 3 to 5 years from having a source of HEU sufficient to be able to manufacture about one nuclear weapon a year (Fainberg, 1993). A smaller, longer-range research and development effort on centrifuge separation of uranium, involving technology obtained from Germany, might eventually have enabled Iraq to achieve a much higher rate of weapon production than is possible with electromagnetic isotope separation (UN/IAEA, 1991). Estimates of the overall budget for the Iraqi program are at least $1 billion annually (Fainberg, 1993). The evidence collected by the IAEA appears to implicate most of the Western nuclear supplier nations in supplying equipment for the Iraqi effort (Muller, 1993; UN/IAEA, 1991).

The Current Safeguards Stalemate In North Korea

An ambiguous nuclear proliferation episode is currently unfolding with North Korea. That nation joined the NPT in 1985 but delayed entering into a safeguards agreement with the IAEA. North and South Korea signed a mutual nonaggression pact in December 1991, agreeing to a nuclear-free Korean peninsula and pledging economic cooperation. North Korea finally signed the safeguards agreement in January 1992, which cleared the way, in May 1992, for the IAEA to make inspections and emplace safeguards equipment at the Yongbyon and other nuclear facilities.

However, a serious dispute arose in February 1993, when the IAEA requested authorization for special inspection of two sites near Yongbyon. These sites were suspected to be undeclared nuclear waste storage depots holding reprocessing waste from a covert effort to obtain plutonium for nuclear weapons. The IAEA had acquired satellite photographs of the two sites and cited evidence from its earlier inspections that North Korea had reprocessed more spent fuel than had been reported to the agency. North Korea denied the IAEA access, claiming the sites were military ones not related to nuclear activities. In March 1993, North Korea announced a withdrawal from the NPT but later suspended the action, while refusing to permit further IAEA activity.

Bilateral negotiations on economic matters and cultural exchange between North Korea and South Korea came to a standstill. In direct bilateral negotiations, the United States insisted that North Korea permit inspection of the two sites. In mid-December, the U.S. Secretary of Defense repeated, in a television program, the U.S. concern that North Korea may have reprocessed, before any safeguards were in effect, spent fuel from a reactor that was shut down for about 100 days in 1989.[10] In February 1994, North Korea agreed to permit IAEA inspections of seven sites where the agency had previously emplaced safeguards equipment.

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[10] See Aspin (1993). The Secretary further remarked that if North Korea did have one or two nuclear bombs, the nation did not appear to be expanding its nuclear capacity.

However, as of now, the issues concerning the two suspect sites are unresolved, as is the prospect for further IAEA inspections.[11]

Yet the creation of a nuclear-free zone on the Korean peninsula, backed by China, Japan, Russia, and the United States, is still a possibility. Such a development could help stabilize relations throughout Northeast Asia and reinforce nonproliferation efforts elsewhere. Conversely, a nuclear weapon capability in North Korea would put pressure on South Korea and Japan to take steps that could destabilize the region and greatly weaken nonproliferation efforts generally. Thus, North Korea may be a more crucial test than Iraq for the viability of nonproliferation in the 1990s.

The Status of Proliferation Barriers and the U.S. Counterproliferation Initiative

The spread of de facto but undeclared nuclear weapon capability underscores the crumbling of the technical barrier to proliferation—over a 40-year period, the knowledge, the know-how, and even the special equipment to produce fissionable weapon materials have become available (see *Assessment of Proliferation Issues for S&T Systems*). Building and operating a major enrichment or reprocessing facility, however, remain formidable. A decade or so seems required in most countries for development and construction. Practical barriers to proliferation could still be effective deterrents if concerned nations show that they will take timely action—political, economic, and, as a last resort, military—given information from satellites, overflights, inspections (where allowed), and intelligence sources.

The recently announced U.S. "counterproliferation" initiative is a major shift in nonproliferation policy (Aspin, 1993; U.S. Department of Defense, 1993). The initiative goes well beyond political and economic efforts meant to deter the acquisition of nuclear weapons or other weapons of mass destruction; indeed, the Secretary of Defense has remarked on the diminished significance of efforts aimed simply at "prevention" of proliferation. The new initiative involves the potential use of military assets against a rogue nation or terrorist group that may have such weapons in their possession. The new policy gives priority to bolstering the ability of the armed forces to respond to the proliferation of weapons of mass destruction in the post—Cold War world—particularly in the former Soviet bloc and in hostile developing countries. The nuclear ambitions in Iran have already attracted congressional attention (Donnelly and Davis, 1992).

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[11] On October 21, 1994, the long and difficult negotiations between the United States and North Korea bore fruit. The two nations signed an agreement under which North Korea would immediately freeze its nuclear weapons program and, over time, take steps to normalize its nuclear activities and facilities. North Korea agreed to allow international inspections in about five years to determine how many nuclear weapons that nation might have produced to that date. In addition, North Korea agreed to dismantle its existing nuclear facilities and to ship radioactive spent fuel rods out of the country beginning in about 10 years. The United States agreed to lift trade barriers and take steps toward diplomatic recognition. The United States also agreed to supply heavy oil to North Korea to make up for the loss of nuclear electrical production. The United States also agreed to arrange for the construction of two 1-GWe LWRs to replace the North Korean gas-graphite reactors. In early November 1994, South Korea lifted its trade ban with North Korea and accelerated discussion of economic issues.

Yet deterring nuclear weapon proliferation requires that the political barriers are credibly expressed in institutions, such as the IAEA, that enjoy the support of the major powers and are acceptable to regional parties. Strengthened IAEA safeguards, under the sanction of a renewed NPT and perhaps buttressed by regional and bilateral agreements and safeguards means, could remain a key cooperative mechanism to forestall the further spread of nuclear weapon capability (Blix, 1992; Jennekens et al., 1992; Scheinman, 1992a; Nye, 1992). This mechanism could be part of a larger initiative on cooperative security in the post—Cold War world that was proposed by two officials of the U.S. Department of Defense (Carter et al., 1992). Counterproliferation means would remain a resort where cooperative means are not successful.

Strengthening Safeguards

The revelations in Iraq, the situation in North Korea, and concerns about problems to come with Iran and Algeria stimulated proposals (1) to strengthen the capability of IAEA safeguards to detect undeclared nuclear facilities and nuclear activities and (2) to codify measures to deal with such facilities and activities when discovered (Blix, 1992; Jennekens et al., 1992; Natio and Rundquist, 1992; Nye, 1992; Fainberg, 1993). Several proposals would make special inspections an established IAEA policy whenever information available to the agency provides a reasonable basis for the action (Blix, 1992; Bunn, 1992; Nye, 1992; Scheinman, 1992a, b). However, top IAEA officials would need much better information, including intelligence data, than has been regularly available. Studies have identified potentially useful technical and nontechnical indicators of covert nuclear activities and facilities (Fainberg, 1993). Another possibility is cooperative assurances that could enhance verification that a nation is not conducting clandestine nuclear activities. Also, the Nuclear Suppliers Group could tighten its procedures and widen its membership to include the emerging nuclear suppliers.

Proposed improvements to deal with suspicious activities and suspected violations center on giving the IAEA more flexibility and authority to respond rapidly, for example, to send in its own inspection team on a few hours notice (Scheinman, 1992a). The IAEA would have to rely on the U.N. Security Council to back such a prerogative. Means to deal with uncooperative nations or proven violations would still depend on embargoes and economic sanctions, as with Iraq and as proposed with North Korea if that nation remains intransigent, and military sanction would be reserved as a last resort.

Another kind of confidence test looms in the near future. Article X of the NPT requires a recommitment to the treaty by all signatories after the treaty has been in force for 25 years, which will occur in July 1995. For years, a lack of substantial progress on reducing nuclear arsenals highlighted the discriminatory nature of the treaty and was a sore point at the 1975 and 1980 NPT review conferences. The end of the Cold War transformed that issue. With the break-up of the Soviet Union at the end of 1991, four nations—Belarus, Kazakhstan, Russia, and Ukraine—were created that had nuclear weapons on their territory, which raised a myriad of problems not yet resolved.[12] Such issues as nuclear security assurances, the reduction of nuclear weapon stockpiles, a cutoff of production of nuclear weapon materials by all nations, and delegitimization of nuclear weapon use are not only important initiatives in their own right but also address the concerns of many nations whose support is essential for the renewal of the NPT (see Scheinman, 1992b; Chrzanowski, 1993).

However, the commitment of the nuclear weapon nations to work toward a Complete Test Ban Treaty, an obligation contained in the Limited Test Ban Treaty of 1963, was a sore point at the review conference in 1990. The recent U.S. policy against testing is an important step but is not a substitute for a treaty involving all the nuclear weapon nations, especially China, which has continued to test. In addition, strengthened IAEA safeguards and their enforcement are now much more serious issues than was the case at previous review conferences.

Recent U.S. Policy on Nonproliferation and the Use of Plutonium

In late 1992, Japanese preparations for the use of plutonium in civil reactors made headlines. Japan shipped about 1.7 tons of plutonium, which was obtained from the reprocessing of Japanese LWR spent fuel in France, to feed Japan's Monju breeder reactor, which is scheduled to start operations in a few years. Because the Japanese spent fuel was of U.S. origin, the Bush administration had given permission for the shipment after obtaining the approval of Congress, as required under the 1987 Nuclear Nonproliferation Treaty. The shipment raised a storm of anxiety and criticism in many nations. Several Southeastern Asian nations even banned the shipment from passing through their waters. Moreover, the Japanese government's plans to make one or two such shipments a year for the next 18 years were not only disquieting to much of the public at large but also to Japanese industrialists who expressed public concern over the financial commitment on the horizon. On February 22, 1994, news reports indicated that the Japanese government decided to postpone, perhaps for as long as two decades, the previous plans for the extensive stockpiling of plutonium and the deployment of several breeder reactors.

With the change of presidential administrations in January 1993, U.S. nonproliferation policy was again the subject of major review, taking into account several factors: (1) the stalemate in North Korea, (2) the lessons from the IAEA special inspections in Iraq, (3) the public response to the Japanese plutonium shipment, and (4) the impending renewal of the NPT. In September 1993, the Clinton administration made a major policy pronouncement on nonproliferation and the use of plutonium: "… the United States does not encourage the civil

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[12] Recently, a study was released by the NAS addressing the risks and issues involved in the disposition of plutonium from nuclear weapons of the United States and the former Soviet Union (see Management and Disposition of Excess Weapons Plutonium, National Academy of Sciences [1994]). The study conclusions emphasize the importance of immediate steps by the United States and states of the former Soviet Union, particularly Russia, to provide effective interim storage of the plutonium from the dismantled weapons and to work toward better protection for public health, safety and the environment, including means for eventual elimination of the material in a manner that meets at least as high a standard as that for the disposition of spent reactor fuel.

use of plutonium and, accordingly, does not itself engage in plutonium reprocessing for either nuclear power or nuclear explosive purposes. The United States, however, will maintain its existing commitments regarding the use of plutonium in civil nuclear programs in Western Europe and Japan" (White House, 1993; Davis and Donnelly, 1993).

The policy statement emphasizes the intention of giving nonproliferation greater priority in diplomacy, and in consideration of regional security and economic matters, and of seeking to promote nonproliferation efforts and making nonproliferation an integral part of relations with countries around the world. The policy also bears on the major U.S. initiative to obtain an extension of the NPT when it comes up for international review in 1995. The plutonium policy discourages any S&T undertaking with LWR spent fuel in the United States in the foreseeable future.

Assessment Of Proliferation Issues For S&T Systems

This section assesses proliferation issues for S&T systems vis-a-vis the LWR once-through fuel cycle. For illustration, the four cases involving the three principal S&T transmutors—the ALMR, LWR, and ATW—are evaluated and reviewed by the committee:

Case 1 is the advanced liquid-metal reactor/integral fast reactor (ALMR/IFR) transuranic burner treated in terms of units comprising nine ALMRs integrated with a local pyroprocessing unit that handles ALMR fuel plus LWR spent fuel transported to the unit. (Centralized aqueous processing is covered in Case 4.)

Case 2 is the "baseline" Los Alamos National Laboratory (LANL) accelerator transmutation of waste (ATW) proposal with heavy-water moderation and onsite aqueous processing for the burning of transuranics (TRUs) and waste from LWRs.

Cases 3 are two versions of the advanced LANL ATW concepts with thorium fuel generating fissile ^{233}U and using onsite processing. One is a self-contained energy producer burning its own waste; the other version burns waste from LWR spent fuel.

Case 4 is a hybrid system comprising ALMRs and once-through uranium-fueled LWRs with processing of spent fuel in large, centralized aqueous reprocessing and fuel fabrication facilities. This case involves plutonium in bulk form at the facilities and in fresh fuel transported to the reactors and is similar to cases evaluated in the 1970s.

The main proliferation issues for the four cases are summarized in Cases 1 through 4 at the end of the appendix. The discussion of issues to follow is organized in terms of three generic components—reprocessing, enrichment, and reactors—which are implemented with different technologies in the four cases.

Reprocessing

Chapter 3 reviews the status of separations technologies and the facility requirements for reprocessing spent reactor fuel. Reprocessing facilities of 1,200-Mg/yr working capacity were used to separate plutonium for weapons at Hanford, Washington, or Savannah River, Georgia. Such a facility could separate perhaps a ton of weapons-grade plutonium per year or some 3 kg per day, depending on the concentration of plutonium in the fuel rods. Thus, 3 days production is enough for one to two nuclear weapons. The UP3 and THORP commercial aqueous reprocessing plants in Europe have a working capacity of 900 Mg/yr each—Case 3 of this appendix uses a reprocessing facility of that type and capacity. For a smaller plant (e.g., 300 Mg/yr) the corresponding time is about a week to separate enough plutonium for one weapon. Existing plants practice materials accountability at about 1% precision with 0.3% to perhaps 0.1% precision claimed to be possible with advanced automated designs. For the plant of 1,200-Mg/yr working capacity, an uncertainty of 1% is enough plutonium for a weapon in a time of about a year. Actual safeguards systems could do better by examining the measurements versus the records for patterns that reveal inconsistencies, and by using techniques for containment and surveillance (Schuricht and Larrimore, 1988). However, some observers conclude that the unaccounted material for a large reprocessing plant may exceed several significant quantities per year (Miller, 1990).

To the committee's knowledge, no existing reprocessing plant had facilitating safeguards techniques as a primary design constraint. In the early 1980s, the Oak Ridge National Laboratory (ORNL) conducted a study of a new facility for reprocessing fuel from the proposed Clinch River fast reactor (Birch et al., 1981). The concept sought to implement all-remote maintenance and accommodate the requirement for safeguards. With the cancellation of the Clinch River reactor, the reprocessing plant design was not carried through to the level of detail needed for construction. A remotely operated and maintained aqueous reprocessing or fabrication plant for bulk mixed-oxide (MOX) fuel—with optimized designs for automated materials accountability and for advanced surveillance and containment techniques and designed to accommodate resident safeguards inspectors at the facility—remains a design goal.[13] However, an ongoing project in Japan for a large, new fuel reprocessing plant, based on aqueous technology, has international safeguards as a major design constraint and might become the first such plant constructed. Whether such a design would significantly affect the evaluation of national diversion scenarios is an unresolved question.

Proponents of pyroprocessing, planned by Argonne National Laboratory (ANL) for the ALMR/IFR system, suggest that the technology lends itself to small, secure facilities, self-contained at the reactor (see Chapter 3). Case 1 of this appendix assumes a system design of this type. An initial assessment has been made of the proliferation implications of a

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[13] A large, remotely operated and maintained MOX fuel fabrication facility designed for safeguards was planned for the early 1990s at Hanau, Germany, to support plutonium recycle in pressurized water reactors. The state of Hesse granted the final license in March 1991, but after much public opposition to the facility, construction was canceled in 1992.

conceptual IFR comprising a fast reactor that uses metallic fuel plus a collocated fuel reprocessing and refabrication plant that uses pyrometallurgical technology (Wymer et al., 1992). The study reached conclusions based on information from research and development on pyroprocessing and on generic considerations of what a commercial-scale reprocessing-refabrication facility would be like. However, the study lacked detailed flowsheets on the process, equipment specifications, and a real plant layout—the kind of data needed for a realistic evaluation of diversion scenarios. The flavor of the study conclusions can be gained from the following excerpt from its executive summary (Wymer et al., 1992:).

> Although it is possible to conceive a variety of ways to remove and purify plutonium from the IFR fuel recycle process with the intention of using it in nuclear weapons, the removal scenarios require off-normal equipment operation or equipment modification and are, for the most part, readily detectable. A large amount of radioactivity remains with the fuel throughout the entire fuel cycle, and virtually all operations must be carried out remotely in inert-atmosphere hot cells. These facts make it difficult to carry out undetected covert operations to divert plutonium to weapons use and provide a substantial period of time between when a country intent on overt proliferation might abrogate the nonproliferation treaty and when it could produce its first weapons-grade plutonium.
>
> The radioactivity of the fission products remaining in the plutonium from IFR, the mix of plutonium isotopes, and the presence of large amounts of uranium make it a less than optimum material for use in weapons production. The radioactivity would make handling and machining of the plutonium and assembly of a weapon very difficult. The relatively high ^{238}Pu fraction would produce heat that would cause phase changes in the plutonium alloy as the temperature rises, resulting in degradation of the performance of the weapon. The heat also could be detrimental to other components in the fabricated weapon during storage.
>
> The intrinsic radioactivity of the recycle plutonium product and the requirement of remote recycle operations in inert-atmosphere hot cells are favorable safeguard factors for IFR. On the other hand, inspectability and material accountability for verification purposes are relatively more difficult for the IFR recycle plant than for a plutonium/uranium redox extraction process (PUREX) plant and a conventional oxide fuel fabrication plant. It is important that the design of the commercial IFR fuel recycle plant give adequate attention to safeguards, inspectability, and accountability features. To provide adequate safeguards likely will require more than customary reliance on containment and surveillance and less reliance on material accountability at key points in the process

This brief excerpt regarding a generic pyroprocessing technology and plant indicates that the process, if practical, may have potential for the future if a realistic and detailed plant design can adequately validate the conclusions. However, pyroprocessing is potentially able to accommodate a wide range of process parameters. Thus, one would want to give further examination to two possible modes of misuse: (1) If new feed material (e.g., low burn-up spent fuel containing little ^{240}Pu) were introduced into an IFR from the outside and run through the system's pyroprocessing equipment, how long would it take to start obtaining plutonium from that equipment and what would be the rate of production? (2) If the normal system operation were altered by shortening the fuel irradiation time to reduce ^{240}Pu concentration in the ALMR spent fuel, could an existing ALMR/IFR system produce weapons-grade plutonium? Further study is needed.

Enrichment

The methods for isotope separation depend on small differences in physical and chemical properties that are due to the slight differences in mass among the isotopes. Four methods have proven practical for production-scale separation of fissionable isotopes and, thus, for enrichment of uranium: (1) gaseous diffusion developed in the 1940s and employed by the United States and the former Soviet Union; (2) gas centrifuge and jet nozzle commercialized in the late 1960s in Europe and also employed by the former Soviet Union; (3) chemical separation developed in France and Japan in the 1980; and (4) laser-induced separation developed in the United States in the 1980s. The less efficient technique of electromagnetic isotope (calutron) separation, developed in the United States in the early 1940s and used to obtain the enriched uranium for the Hiroshima bomb, was adopted by Iraq for its covert uranium enrichment program.

Safeguards are challenging to employ effectively at a large enrichment facility. Consider, for example, an enrichment plant with a capacity of 1 million separative work unit/year (SWU/yr) using gaseous diffusion or centrifuge technology. Although some commercial plants are five to ten times larger, 1 million SWU/yr is typical of many operational facilities and is convenient for estimates. Numerous separative stages are required in such an enrichment plant, because each stage achieves only a small fractional isotope separation. Each stage typically consists of many individual units connected in parallel. Stages are cascaded in series by means of piping to achieve whatever degree of enrichment is desired. For enrichment at 0.2% tails assay, 236 SWU are expended per kilogram of 93% HEU produced, whereas to produce 3% LEU, 140 SWU are expended per kilograms fissile material or 4.3 SWU are expended per kilograms of 3% diluted fuel feed. Thus, the nominal 1 million SWU/yr plant configured for low enrichment from natural uranium feed material could produce about 20 tonnes of 3% LEU fuel feed material per month or 240 Mg/yr, which could serve the needs of about eight LWRs of nominal 1,000 MWe capacity.

Alternatively, the same plant reconfigured for HEU production—reconnecting units to configure more stages in series—could produce about 350 kg of HEU per month or enough nominally for about a dozen nuclear weapons per month. For centrifuge plants, which are much more readily reconfigured than diffusion plants, safeguards procedures were worked out in the 1980s by a working group and applied at plants in Europe (Menzel, 1983). Such safeguards employ tamper-proof seals and surveillance equipment on all entrances to the plants operating areas to prevent the reconfiguration of the stages, with sampling of output and waste material for assay on regular inspections every few weeks and on random spot inspections.

The chemical separation approach is reportedly capable of substantial enrichment (Mikake et al., 1984). With many stages, it could produce HEU. A facility for the process is much like a conventional chemical processing plant. If such a plant is operated under safeguards with resident inspectors, then the risk that a modification for HEU production could go undetected may be low. However, much of the separative work is expended in getting to 3.5-4% LEU, so safeguards must also be applied to the LEU output and overall material balance. Otherwise, material diverted from a chemical separation plant (or centrifuge plant) producing LEU could be further enriched in a small, covert "topping" plant, using the same or another enrichment technique, to achieve HEU from the combination. The topping plant itself might be hard to locate without some intelligence information.

Recently, laser isotope separation has been developed in the United States for purifying plutonium (i.e., separating ^{239}Pu from other plutonium isotopes) and for enriching uranium. The process lends itself to a plant of modest industrial scale and was selected by the U.S. Department of Energy (DOE) in the mid-1980s as the technology of choice for enrichment in the United States. The high-power laser technology is very high-tech and may be difficult for many developing countries to acquire in the near term. However, other laser-induced chemistry applications would use much the same type of lasers, which may give impetus to their development and widespread commercial use. If so, it is likely that technology to practice the laser isotope technique would become generally available over the next decade or two.

The application of these considerations to the S&T cases depends on whether the case merely requires HEU for start-up or a periodic supply of LEU for make-up to support operating LWRs, as in Cases 1 and 3. For start-up on HEU, the reactors presumably could draw down existing HEU stocks, which could be augmented if necessary by a special campaign conducted in commercial-scale enrichment plants that exist to support the current once-through fuel cycle. Then those enrichment plants could be shut down or mothballed. A case requiring ongoing LEU make-up requires operating enrichment plants, albeit under safeguards. Of course, the perceived proliferation risk would depend strongly on where enrichment plants are located and the institutional arrangements that prevail. The Argentina–Brazil bilateral safeguards agreement covering their enrichment plants, which is backed up by IAEA inspection, is an example of a multilateral institutional arrangement that might be satisfactory in other instances (Redick, 1990; Krasno, 1992). Even then, abrogation scenarios (a nation renouncing a treaty or agreement and seizing the plant) would remain a potential proliferation risk (see the section *Proliferation Issues* in Chapter 6).

Reactors

The first sections of this appendix briefly review the LWR once-through fuel cycle as the proliferation baseline and introduce two other types of thermal reactors—the HWR and the LGR. Reactors moderated with either heavy water or graphite permit on-line refueling, as discussed below. They were used for weapons-grade plutonium production by the major nuclear-weapon nations in the past; a few nations continue to do so. Chapter 4 reviews the LWR in detail and also reviews fast reactors, such as the ALMR, that can operate efficiently as breeders.

A major class of national diversion scenarios entails diversion of spent fuel at a reactor site. A large reactor is not a trivial matter to safeguard. Reactor-grade plutonium is produced at a rate of about 0.9 of plutonium per MWt·day or 0.33 kg MWt/yr per MWt·year. Weapons-grade plutonium requires a shorter irradiation time than reactor-grade to suppress production of ^{240}Pu and may need a greater fraction of the year for loading/unloading operations. For example, a 2,200 MWt reactor operating at 80% of nominal capacity would produce about 600 kg of reactor-grade plutonium per year. Such a reactor would produce less, for example 300 to 450 kg of weapons-grade per year depending on whether the reactor could be refueled on-line (i.e., a change of fuel rods by a machine without shutting down the reactor) or would require batch refueling involving a shutdown. 300 kg of plutonium per year is material for 40 to 60 weapons per year or 3.5 to 5 weapons per month.

IAEA safeguards defend against the diversion of spent-fuel rod bundles with tamper-proof seals and tags (Fischer and Szasz, 1985). Safeguards inspectors install these devices as the fuel rods are placed in the reactor. The tags are checked when the spent fuel is removed from the reactor for placement in the storage pool, and they are checked again in the pool on each inspection. The safeguards technologies also include gamma spectral measuring equipment and surveillance instrumentation. A reactor moderated with ordinary water (a LWR) normally requires batch refueling during a shutdown, which provides safeguards inspectors with an opportunity to be present while fuel is being loaded/unloaded. Inspectors can also make regular and randomly timed visits to check seals and tags on spent fuel in storage as well as read other safeguards instruments and check operating records to verify that there has been no unannounced shutdown of any consequence between scheduled shutdowns for refueling and maintenance.

Reactors for which on-line refueling is possible pose a severe challenge to a safeguards regime. The Russian RBMK-type graphite-moderated reactor, for example, uses a machine that moves progressively over the top of the core on a schedule to remove irradiated rods and insert fresh ones. The CANDU, an HWR, uses a similar machine for its core in which the fuel rods lie horizontally. Such reactors require more extensive safeguards than batch-refueled LWRs. The CANDU safeguards might serve as a model (Zarecki and Smith, 1981; Ferraris and Wredberg, 1982). Safeguards instrumentation uses core input and discharge counters and monitors the progress of the refueling machine with sealed surveillance equipment through the reactor's upper observation port. All rods entering or leaving the reactor are subjected to inspection, and reactor operating conditions are monitored with sealed detectors to substantiate the operating logs.

Fast reactors present yet another safeguards issue. In addition to fuel rods in the reactor core that contain plutonium, a fast reactor has blanket regions for breeding ^{239}Pu in rods that contain ^{238}U (natural or depleted uranium) and that could be changed while the reactor is operating (on-line). The blanket would be a special target in diversion scenarios that include covert substitution of blanket roads, which are taken away and reprocessed to recover the ^{239}Pu. Thus, fast reactors would need an operating regime with an optimized design to facilitate containment and surveillance safeguards and a design to minimize the possibility of a covert change of blanket rods. Finally, a new safeguards issue is raised with reactors that use fluidized fuel, like the ATW; there are no fuel rods to count! No safeguards scheme has been devised for such reactors.

Summary of Proliferation Issues for the Four Cases

Several STATS committee proposals would significantly change the present situation with the once-through LWR fuel cycle. Case 1 and Case 3 introduce fast reactors as transuranic (TRU) burners in which the core and blanket contain rods with TRU waste. Case 1 also uses fast reactors as plutonium breeders and has a pyroprocessing facility collocated on each site. Both cases require safeguards against covert on-line refueling and against substitution of blanket rods containing uranium for those containing TRU waste. The diverted rods could be reprocessed using the collocated pyroprocessing equipment or taken away and reprocessed elsewhere. The ALMR/IFR proposal has not presented a safeguards regime, so no detailed assessment is possible. Case 3 uses large centralized aqueous reprocessing and fuel fabrication facilities containing plutonium in bulk quantities and transports fuel rods containing plutonium to and from the reactors and the fuel processing sites. Thus, Case 3 raises plutonium diversion issues, as discussed above under *Reprocessing*.

The ATW fludized fuel, a heavy-water slurry in Case 3 and a molten salt in Case 3, which circulates in the reactor proper and the onsite reprocessing unit, requires a new safeguards regime, as noted above under *Reactors*. In Case 3, timing the reprocessing of the protactinium to avoid ^{232}U (which decays to highly radioactive daughters) would enable the production of nearly pure ^{233}U bomb material. In addition, the ATW would be an excellent source of neutrons for covert plutonium production in natural or depleted uranium. For example, if neutrons from the target were used with no further multiplication, the neutron flux would still be about one-twentieth of the normal flux for an ATW with a K_{eff} of 0.95 and would have a hard spectrum.

To minimize the possibility of either covert plutonium production with target neutrons or diversion of either fluid fuel or separated processing streams, an ATW may require sealed entrances to the facilities and extensive monitoring that has continuous surveillance equipment and secure communications and perhaps also resident inspectors like an enrichment facility. The ATW proposal does not discuss safeguards requirements, so no detailed assessment is possible of the effectiveness of such safeguards. LANL has not assessed the feasibility of materials accountability safeguards with the intense radiation field of the fluid fuel (see Chapter 3).

Finally, there is another class of proliferation scenarios beyond diversion scenarios addressed by IAEA and other safeguards, namely abrogation scenarios, which involve seizure of facilities and stores that are under safeguards by a nation after treaty withdrawal or abrogation, followed by the rapid production of nuclear weapons (see Chapter 6). The S&T fuel cycles in general, and the four cases of this appendix in particular, introduce a critical, qualitative change relative to the once-through LWR fuel cycle. That change is the availability of facilities that could be seized for HEU or plutonium production and in which reactor-grade plutonium is normally available in bulk form. As noted in the section *Proliferation Issues* in Chapter 6, abrogation scenarios raise mainly political issues, although technological considerations can be important in determining the time scale in which political and military responses by other nations could limit the production of the nuclear devices. Of the four cases, Case 4 with large centralized facilities and bulk stores, is perhaps the most vulnerable to abrogation scenarios, because the time scale for response by other nations would be the shortest.

Case 1: Integral Fast Reactor Breeder/Burner [14]

The units comprise nine ALMR modules integrated with a local pyroprocessing unit that handles ALMR fuel plus LWR spent fuel transported to the unit to be reprocessed. Any LWRs are assumed to be phased out as they reach the end of their operational life. They are replaced by ALMRs operating as breeders to provide the additional make-up for the waste-burner ALMRs. As accumulated LWR spent fuel is exhausted, ALMRs switch operation to breeder/burners.

Reactors: Nine 155-MWe ALMR modules. Waste-burner ALMRs operate with a breeder ratio of 0.65. ALMRs shift to a breeder ratio near unity to breed plutonium after the LWR waste is consumed. ALMR core and blanket rods could be substituted covertly to make low burn-up plutonium for weapons.

Enrichment: (a) Not required for ALMRs if start-up is on plutonium recovered from spent fuel. (b) For start-up on HEU, ALMRs could draw from existing supplies, augmented as necessary by a special campaign in existing enrichment facilities under safeguards. (c) 1 million SWU/yr per 8 operating LWRs until all LWRs are phased out.

Fuel Reprocessing and Refabrication: (a) ALMR fuel processing of 7.6 kgMg/d (2.8 MgHM/yr) capacity local to the reactor site, using pyroprocessing technology. (b) Additional LWR fuel processing that uses onsite pyroprocessing with a different front end; extra capacity depends on the desired rate for reprocessing the accumulated LWR spent fuel. (Alternatively, a large aqueous reprocessing plant and an associated fuel fabrication plant could be used for LWR spent fuel, as discussed in Case 4).

Spent-Fuel Storage: Temporary storage onsite.

Transportation: (a) Initial plutonium-or HEU-bearing fuel material for ALMR start-up. (b) For ALMR-burners, periodic shipments of plutonium-bearing LWR spent fuel is required; in addition, ALMR fuel occasionally may be transported between multireactor sites to maintain balance.

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[14] Adapted from Argonne National Laboratory ALMR/IFR proposal.

Waste Disposal: Temporary storage and permanent geological isolation of plutonium-bearing waste.

Case 2: Baseline Accelerator Transmutation Of Waste [15]

This is the "baseline" LANL ATW with onsite aqueous processing, which delivers about 1,600 MWe, to the electrical grid. Fluid fuel, consisting of a slurry of tiny particles in heavy water, is used to burn the TRUs, radioiodine, and technetium from accumulated LWR spent fuel.

Reactors: Accelerator-driven neutron-generator-multiplier assembly comprising a 1,600 MeV 250 mA proton accelerator with a tungsten target to produce neutrons that are multiplied in a heavy-water moderated assembly with $K_{eff} = 0.95$. The system produces about 8,300 MWt total power burning the waste from past and existing LWRs containing TRUs and iodine and technetium fission products. The ATW neutrons could be used covertly to make low burn-up plutonium for weapons.

Enrichment: (a) Not required for an ATW if start-up is on plutonium-based spent fuel. (b) For start-up on HEU, ATW could draw from existing supplies, augmented as necessary by a special campaign in existing enrichment facilities under safeguards.

Fuel Reprocessing and Refabrication: (a) Fluid fuel reprocessing of 55 kgHM/day (20 MgHM/yr) onsite integrated with refabrication using aqueous technology. (b) Reprocessing of LWR spent fuel in a large facility (300 Mg/yr) with integrated fuel fabrication, to provide fissile make-up for the ATW.

Spent-Fuel Storage: Temporary pool storage onsite.

Transportation: (a) Initial plutonium-or HEU-bearing fuel material for ATW start-up. (b) Periodic shipments of plutonium-bearing LWR spent fuel.

Waste Disposal: Temporary storage and permanent geological isolation of plutonium-bearing waste.

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[15] Adapted from Los Alamos National Laboratory ATW-1 proposal (1992).

Case 3: Advanced Accelerator Transmutation of Waste [16]

Consist of two versions of the LANL advanced ATW using fuel dissolved in a molten salt with onsite processing. The systems use thorium to generate fissile ^{233}U and deliver 2,180 MWe to the electric grid. One is a self-contained cycle burning its own waste; the other version also burns LWR waste.

Reactors: Accelerator-driven neutron-generator-multiplier assembly to produce neutrons comprising a 800 MeV 110 mA proton accelerator with a molten-lead target that are multiplied in a graphite-moderated assembly with K_{eff}=0.95. The system produces about 6,000 MWt total power in either version. The ATW neutrons could be used covertly to make low burn-up plutonium for weapons.

Enrichment: (a) Not required for an ATW if start-up is on plutonium-based spent fuel. (b) For start-up on HEU, ATW could draw from existing supplies, augmented as necessary by a special campaign in existing enrichment facilities under safeguards.

Fuel Reprocessing and Refabrication: Fluid fuel reprocessing on-site using a nonaqueous technology integrated with fuel refabrication. Version 1: 9.8 kgHM/day (3.6 MgHM/yr); version 2: 8.2 kgHM/day (3 MgHM/yr).

Spent-Fuel Storage: Version 1: temporary pool storage onsite.

Transportation: Version 1: periodic shipments of plutonium-bearing LWR spent fuel; version 2: no fuel transportation.

Waste Disposal: Temporary storage and permanent geological isolation of plutonium-bearing waste.

Case 4: Hybrid Almr/Lwr System [17]

The system consists of groups comprising 16 ALMR TRU burners and 30 LWRs burning technetium and iodine fission products, with aqueous reprocessing of both fuels in large central facilities with associated fuel fabrication. The fuel rods are transported to and from a large number of reactors. This case contrasts with Case 1 in that here the LWRs remain operational, supplying TRU make-up to the ALMRs; also, the reprocessing is centralized.

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[16] Adapted from the Los Alamos National Laboratory ATW-2 and ATW-4 proposals (1992).

[17] Adapted from the Westinghouse Hanford Company and Battelle Northwest Laboratories CURE proposal.

Reactors: (a) 400-MWe ALMRs that operate as waste burners with a breeding ratio assumed to be 0.8 in the CURE proposal. (b) 1,000-MWe LWRs to burn some waste, supply make-up for the ALMRs, and generate electrical power.

Enrichment: (a) Not required for ALMRs if start-up is from reprocessed spent fuel. For start-up on HEU, ALMRs could draw from existing supplies, augmented as necessary by a special campaign in existing enrichment facilities under safeguards. (b) 3.5 million SWU/yr required for each 30 operating LWRs.

Fuel Reprocessing and Refabrication: Central reprocessing facility of 750-Mg/yr working capacity per group using aqueous technology, with fabrication of 1,000 fuel rods/yr per group from input of 300 Mg/yr of uranium and 20 Mg/yr of plutonium.

Spent-Fuel Storage: Temporary pool storage onsite.

Transportation: (a) Initial plutonium-or HEU-bearing fuel material for ALMR start-up. (b) Periodic shipments of plutonium-bearing fuel between sites and centralized fuel-processing facility for the operating LWRs.

Waste Disposal: Temporary storage and permanent geological isolation of plutonium-bearing waste.

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Appendix I

Health And Safety

Introduction

When discussing health and safety issues, it is important to be precise about the meaning of the terms involved. Three terms used extensively throughout this report are "safety," "risk," and "hazard." In general usage, they are used with slightly different meanings. Safety is the catchall descriptor having the meaning found generally in most dictionaries, i.e., the condition of being safe from undergoing or causing hurt, injury, or loss. That is, it prevents a negative consequence directly related to human health and well being. The word "hazard" is also used in the sense of its usual dictionary definition, that is, as a "source of danger." A hazard may produce a negative consequence under certain scenarios, but the presence of a hazard is not necessarily a risk to the public in and of itself. The problem is that the word "risk" is often used to mean hazard and safety when, in fact, in the nuclear business in particular, it has quite a different meaning. For this report, the committee has adopted the triplet meaning of risk (Kaplan and Garrick, 1981) that is widely used in the probabilistic risk-assessment community. In particular, risk assessment answers three basic questions:

1. What can go wrong?
2. How likely are things to go wrong?
3. What are the consequences?

The first question is answered by structuring scenarios (sequences of events) of the different ways that a facility can get into trouble. This step of risk analysis requires a clear understanding of how the facility operates, the hazards involved, those events that can initiate an accident scenario, and how the facility can fail to respond safely.

The question of likelihood is often a matter needing detailed analysis, since, in many instances, there are too few data to assign statistically based frequencies. The most common approach is to develop frequencies and probabilities based on all available evidence, including detailed logic models that decompose the facility into components for which there are data and increased knowledge, and to do so in such a way as to recognize the uncertainties involved.

Finally, the consequences are defined in terms of injuries, fatalities, facility damage, environmental damage, etc., or combinations thereof. There may be interest in a variety of types of consequences. Thus, a risk assessment is a structured set of scenarios, their likelihoods, and their consequences. The results not only provide information on the likelihood of different levels of severity but also convey the analysts' confidence (uncertainty limits) in the results.

In this report, when the discussion is about scenarios, likelihoods, and consequences, generally the term "risk" is used. The term "hazard" most often refers to the nature of the materials and inventories involved, including their toxicity, but not to likelihoods and consequences of upsets and accidents that get the materials into the biosphere. Safety is used in a qualitative and much more general sense.

When applied to the radioactive waste problem, environmental, health, and safety issues are closely intertwined. The health effects of concern are the negative consequences of being exposed to ionizing radiation. By far the major concerns are latent cancer and genetic effects from low doses, although accidents during the transmutation, separation, and handling phases have some potential for producing somatic effects. The physical principle involved is that a person will receive a dose only when radioactive material is in relatively close proximity and unshielded. Environmental impacts are generally associated with contamination of soil or ground water that could result in an eventual transport of radioactive material and exposure of the population, unless there is some type of isolation (resulting in loss of land use) or an extensive cleanup effort to prevent the gradual spread of the contaminants. Safety concerns refer to near-term scenarios that breach the engineered facilities to produce the resultant environmental contamination or more direct (e.g., airborne) pathways of radioactive materials to workers or the population.

The likelihood that a set of conditions will actually occur that could produce a dose that results in health effects can be estimated with a risk assessment of the scenarios that produce those conditions.

This section highlights the risks and safety issues associated with the various concepts being proposed for the reduction or alteration of radioactive waste. The information presented is based on work performed by the proponents of the various concepts. While there is no attempt to perform an independent analysis to verify the details of the claims of the proponents, there is an attempt to provide a limited, objective review of the risk and safety work performed to date.

The approach taken is to develop risk and safety summaries of each of the radioactive waste treatment concepts based on studies and reports made available to the Committee on Separations and Transmutation Systems. These summaries are at the end of this appendix. The risk and safety summaries all have the same format to facilitate comparisons of the different concepts and serve as the centerpiece for the discussion and conclusions that follow. The radioactive waste treatment concepts are identified to correspond with the major concepts described in this report and generally are as follows:

- reactor-based

- —advanced liquid-metal reactor (ALMR)/integral fast reactor (IFR),
- —light-water reactor (LWR),
- —particle bed reactor (PBR);

- accelerator-based

- —Los Alamos National Laboratory (LANL) accelerator transmutation of radioactive waste (ATW),
- —Brookhaven National Laboratory (BNL) Phoenix;

- Clean Use of Reactor Energy (CURE; complementary mix of concepts); and
- base case (LWR once-through cycle).

Seven issues are presented in the attached risk and safety summaries to highlight factors that would assist in the selection of a potential mix of facilities from a safety and health risk point of view. The first group of issues summarizes the status of evidence regarding the overall concept presented by each proponent. The issues include the systems technology maturity, quantitative safety assessments, and the experience demonstrating feasibility. As a general rule, the greater the uncertainty regarding the system design, interfaces, and performance under actual operating conditions, the higher the likelihood of unforeseen functional and phenomenological events, that is, the higher the risk. The second group of issues lists the selected design features in terms of applicability to waste objectives, unique risk reduction features, unique safety concerns, and major sources of uncertainties. These issues highlight the ability of the technology to address the risks associated with permanent geologic disposal, the advantages and disadvantages of the design with respect to risk, and unresolved issues to ensure safety.

Because of the great differences in the nature of the radioactive hazard from step to step in the fuel cycle, the seven issues are addressed for each of three phases common to all concepts: transmutation, separation, and storage.

During the transmutation process, the safety concerns are very similar to those normally associated with the operation of nuclear reactors. Except for the one accelerator concept involving a homogeneous blanket, the radioactive material is contained in either solid fuel or target material. For the LANL ATW concepts, the fuel is already in a liquid and mobile state in the event of any kind of containment failure. Similar to nuclear reactors, the accident pathway of greatest concern is a release into the atmosphere. This is primarily because of the power levels, temperatures, and decay heat involved. Pathways other than atmospheric dispersion are believed to be less important in this phase because the mobility of molten material (corium), once it leaves the immediate region of the core, will be severely restricted as it solidifies within the confines of the facility, where it could be recovered. As with commercial nuclear reactors using the once-through fuel cycle, the basic approach to safety is defense-in-depth with both active and passive safety systems. Unlike commercial nuclear reactors, there is a lack of operating experience with any of the advanced reactor or accelerator-based transmutation concepts.

The separation phase presents a different set of challenges. Although the high power levels and temperatures of the transmutation phase no longer are present, the material is expected to be sufficiently radioactive to generate significant quantities of heat. Moreover, to facilitate separation, the material will be converted into liquid form during appropriate parts of the separation process and transported throughout a more dispersed facility. Thus, although the driving force for airborne dispersal has declined, the physical form of the materials is more conducive to widespread contamination. Finally, since the material will be actively handled over a larger facility, maintaining tight control and defense-in-depth at all times will be more difficult.

For the disposal phase, the concern is the eventual degradation of the engineered barriers and the uncertainty in the ability of the geologic formation to keep the longer lived isotopes isolated from the biosphere. Once the material is packaged and placed in the repository, and once the repository is sealed, there is a loss of active control over its mobility. The acceptability of the repository then hinges on society's confidence that enough has been done to maintain isolation of the waste long enough to prevent health and environmental effects.

Discussion of Concepts

All of the proposed concepts have some health and safety characteristics in common. First, as their objective is to reduce the requirements for a geologic repository, they all produce a reduction in long-term risk. Second, as they all require some form of spent-fuel processing, the concepts all present a short-term risk during the separation phase that is currently bypassed by the once-through fuel cycle. Conversely, to the extent that they produce a net positive energy output through the use of recycled actinides, the concepts all have the potential to reduce the short-term risk due to uranium mining and milling operations. Third, all transmutation technologies have the objective of providing some form of a passive safety feature not employed in currently licensed LWRs to reduce the likelihood of the release of radioactive material into the environment. Consequently, the proponents claim at this conceptual stage that an integrated separations and transmutation (S&T) system will produce a net decrease in health and safety risk to the public. As discussed later, there is insufficient evidence to support this claim at this time.

Before addressing the impact of an integrated S&T system on risk due to high-level waste, it is reasonable to compare it with the front end of the cycle to determine if resources are being focused on that aspect of the problem that would provide the greatest risk reduction. The mining and milling operations disturb the geologic formations that retain naturally occurring radioisotopes and allow some public exposure. Moreover, the depleted uranium tails can produce a continuous source of radon if reasonable disposal methods are not employed. In the long run, the main concerns are ^{226}Ra and ^{210}Pb (from the decay of ^{238}U) and ^{231}Pa, a daughter product of ^{235}U. Michaels (1992, and private communication, 1993) of the Oak Ridge National Laboratory has presented summaries of potential health risks (effects) among the total U.S. population for the current once-through LWR fuel cycle and for an ALMR fuel cycle. The potential for radiation exposure from mining and milling operations is estimated to be approximately 50% of the total exposure from the existing nuclear fuel cycle, and the waste management exposure potential is about 1% of the total exposure potential.

Comparison of the risk implications of selected design features of the various S&T concepts is difficult primarily because of the differences of the specific objectives of the individual technologies. Two of the concepts, the accelerator reactor combinations and the PBR, have been conceived specifically to transmute wastes. Therefore, they have some revolutionary design features that provide unique opportunities to reduce the risk of long-term high-level waste disposal. However, these concepts introduce near-term operational safety concerns that have not yet been resolved. There remains the problem of the low-level waste (LLW) and its risks, which could be significant by comparison. Except for the Phoenix concept, all provide a mix of reactors and reprocessing facilities to make effective use of the energy-producing capabilities of the actinides, with the important by-product of also reducing the quantity of high-level wastes (HLW) that must be geologically isolated. These concepts tend to be more evolutionary; they do not eliminate the requirement for disposal but simply reduce the magnitude of the disposal.

Based on the information in the summaries and discussions at the end of this section, it can be seen that the accelerator concepts are still in a very preliminary conceptual stage of development, with little or no actual experience yet available to support the development process. The PBR has some basis in existing high-temperature gas-cooled reactor (HTGR) technology, but the proposed design parameters for the PBR concept go far beyond the HTGR experience. The fast reactors have progressed further into the development process, and it appears that many of the technologies have been demonstrated, at least experimentally or in a prototype. LWRs are by far the most developed of any reactor type, and many of the recent designs can be modified to become efficient plutonium burners. The ALMRs offer even more advantages in terms of passive safety systems. The uncertainties with the use of LWRs center around the more stringent reactivity control requirements of mixed actinide fuels. The CURE concept is not tied to any one design philosophy, and so it cannot be compared directly with specific reactor types. Rather, it selects a mix of the above technologies and gradually integrates them over a long development process to optimize both energy production and reduction of long-term waste disposal requirements. The CURE concept, if properly followed, would tend to reduce the risks associated with problems that arise from a commitment to one technology about which there is little information.

ATW

The proponents of ATW as a waste treatment concept point to several safety advantages of the approach. Most notable are that ATWs operate below nuclear criticality, reduce the source term of radioactive material by continuous processing of the liquid fuel, and can transport liquid fuel to safe geometries for decay-heat removal. The proponents state that the ATW accelerator controls can rapidly shut down on any system malfunctions. When combined with continuous fission product removal, this feature will reduce the heat load on the safety systems following an initiating event.

Among the safety concerns are the ability to maintain system integrity at the beam/target/blanket interface, the reduction in defense-in-depth with the fuel already in a liquid state, and the fact that the accelerator's localized particle beam can initiate a variety of events. Activation of replaceable components, such as beam targets, may even increase short-term storage requirements.

It should be noted that continuous processing does not totally solve the safety problem with respect to fission product inventory. The processing rates will not be sufficiently rapid to mitigate the most severe shutdown heating transient on loss of normal cooling. However, the short delay times between transmutation and separation will result in much higher activities of short-lived fission products in fuel processing equipment. Reactivity swings are another issue that will have to be studied. Fluid fuel reactors typically have very large negative temperature coefficients of reactivity. Therefore, a large reactivity swing can be expected in going from hot operating conditions to cold shutdown. The nonaqueous ATW that is fueled partly or completely with thorium will be subject to a reactivity transient safety issue at high neutron flux.

There are questions about the homogeneity and chemical consistency of the blanket that could impact fuel mobility along its flow path and produce the possibility of hot spots. There are a number of questions that have to be answered about system behavior during both abnormal and transient conditions.

The ATW approach to the treatment of radioactive waste is handicapped by a lack of experience and therefore knowledge about safety issues. No significant and quantitative risk assessments have been made on any phase of the ATW concept, much less the integrated system. Thus, the state of knowledge about ATW safety is not such that an independent confirmation can be made of the safety claims of its proponents.

Accelerator (Phoenix)

The Phoenix accelerator concept has some of the same safety advantages as ATW, most notable is the subcritical mode of operation. It employs a large linear proton accelerator with solid oxide targets in a configuration resembling the core of the Fast Flux Test Facility (FFTF) reactor, so separation can build on established processing technology. However, the pre-and postirradiation processing schemes have not been developed. The specific transmutation targets are neptunium, americium, curium, and much of the iodine produced by LWRs. The direct impingement on the target material, together with the fact that it would operate with a fast neutron spectrum, allows use of some known technologies.

There are a number of uncertainties with this concept. One is the behavior of the target material under combined proton and neutron irradiation. There may be safety issues associated with a failed beam raster, for example, burning holes in targets with the attendant radioactivity release. And decay heat is an issue even with a rapid removal of the proton source.

The most significant shortcomings of the concept are the absence of a design and the lack of any in-depth safety analysis. There is insufficient information to reach definitive favorable conclusions on the safety of the Phoenix concept.

ALMR/IFR

The ALMR/IFR approach to the reduction of actinides has been under consideration for many years. The technology of fast reactors is reasonably well established, at least much more so than for waste reduction concepts based on the use of accelerators. EBR-II has been operating since 1963, and numerous other prototype fast reactors, including the FFTF, have accumulated a substantial experience base. The result is a considerable amount of knowledge on the safety of fast reactors. Some of the safety advantages are the large heat capacity of the reactor coolant system and the possibility of achieving safe shutdown independent of active systems.

Even with the substantial amount of fast reactor experience, there is still little experience on a commercial scale. The fast reactor experience to date has been largely with experimental and prototype special-purpose reactors. While EBR-II and FFTF experiences have been favorable, there have been difficulties with some fast reactors. Specific examples are fuel damage in the Fermi reactor and the EBR-1, cracking of primary system components in a U.K. fast reactor, and steam generator leaks in France's Phoenix reactor and the former Soviet Union's BN-350.

Unlike the accelerator concepts, there have been considerable safety analyses performed on fast reactors. For example, a probabilistic risk assessment (PRA) performed on EBR-II has produced results that are more favorable than most LWRs. There has also been a PRA performed on the ALMR with favorable safety results. However, these risk models have not been subject to the intense reviews that are typical of licensed commercial nuclear power plants. Furthermore, the scopes of the PRAs of fast reactors and LWRs do not appear to be comparable. For example, the PRA results of LWR indicate the extreme importance of the dependencies of safety functions on support systems and site-and plant-specific considerations. Contemporary LWR PRAs also include extensive consideration of human reliability and detailed studies of long-term accident recovery capability. Before a favorable conclusion on the risk of ALMR/IFR concepts could be reached, scope comparisons would have to be made of the different PRAs. PRAs that have been performed on fast reactors have not been scoped against a specific design that has evolved for the explicit purpose of transmuting radioactive waste.

Separation technologies to support ALMR fuel cycles are in a less mature stage. it is anticipated that pyroprocessing will satisfy this need, and pilot-plant studies have been initiated. However, it has not yet been demonstrated at a scale required to support an ALMR fuel cycle. While ALMR/IFR technology and its associated safety assessment are much further developed than for accelerator-based concepts, uncertainties remain on the safety of waste reduction processes using fast reactors.

LWR

The safety of LWRs has the least uncertainty of any reactor type. The present generation of nuclear power plants is dependent primarily on active systems to mitigate accidents, should they occur. The reliability of those systems has been analyzed extensively, and their performance characteristics are well understood. In addition, newer evolutionary designs, including the ALMRs, enable safe cold shutdown with passive systems. These designs are being subjected to PRAs as part of their certification process. However, the large experience base with LWRs, at least for power applications, is with low enriched ^{235}U fuel. There are safety issues, such as more stringent reactivity control requirements, that would have to be studied when using mixed actinide fuels in LWRs.

There is a high state of knowledge on fuel reprocessing from experiences gathered during the initial years of nuclear fuel-cycle development, at least in terms of the plutonium and uranium recovery by extraction (PUREX) process, in spite of the fact that reprocessing of commercial fuel in the United States ceased many years ago.

With respect to safety, the biggest difference in the safety area between LWR-based waste reduction applications and other proposed concepts is that there is very strong and quantitative evidence on the safety of LWRs and little to no evidence on the other concepts. In the end, it may be possible to demonstrate others to be more safe but not on the basis of the evidence currently available.

PBR

The particle bed reactor has been proposed by the BNL as a transmutor for the minor actinides, some of the plutonium, and long-lived fission products from LWR spent fuel. Little is known about the safety of the PBR, as the entire reactor concept is very preliminary. There are some safety concerns. The very high power density (5 MW/liter) make it possible to transmute a significant fraction of actinides, thus eliminating the need for geologic disposal. However, this may also make it difficult to ensure that, on loss of coolant flow, an adequate means of core cooling can be provided in time to prevent excessive heating of core materials. The delivery of high-speed helium coolant (275 m/s) at a reasonable pressure drop remains to be validated at steady state. Also, the effects of burn-up reactivity swings on the operations of such a reactor remain to be evaluated, together with the power distribution issues characteristic of high thermal-flux systems. Even though no significant safety studies have been performed, the proponents of the PBR believe that many key technologies have been demonstrated by the experience of the HTGRs, despite the fact that power densities for the current graphite reactors are more than two orders of magnitude lower than those proposed for the PBR. Finally, the technology for reprocessing the PBR fuel has not yet been demonstrated.

There does not yet exist a technical basis for concluding that a PBR-based waste reduction facility could be operated safely.

CURE

CURE is a concept for mixing existing thermal and fast reactor technologies in some economical way to achieve a complex of both power-producing and waste-reducing nuclear facilities. The result is that the risk and safety issues are very dependent on the mix of reactors actually selected. However, it is reasonable to conclude that more is known about the safety of the CURE approach than about accelerator-based approaches, that is, there is a great deal more known about the safety of the reactor types involved in the CURE concept than about the safety of the accelerator systems being proposed.

Base Case

The base case is the current open fuel cycle of permanently storing the spent nuclear fuel and other radioactive wastes in geologic repositories and allowing them to decay at their natural half-life. From a risk and safety standpoint, there is a minimum of waste handling and treatment prior to permanent disposal. Consequently, the base case represents less near-term risk than any of the S&T systems, all of which require active handling of the radioactive waste in the short term. However, if the once-through cycle is used exclusively in nuclear power plants, the base case does present greater long-term risk during the permanent disposal phase, as well as more risk due to mining and milling operations. There is considerable evidence that the health consequences from permanent disposal are extremely small, although the uncertainty of such postulated consequences is relatively high because of the long time periods involved.

Conclusions

Risk assessment is a matter of quantifying our state of knowledge about the threat to society of a particular system, engineered or otherwise. Generally, systems (that exist only on paper) involving hazardous materials and then only in conceptual form are on the high end of the risk spectrum due to a poor state of knowledge about possible accidents and consequences. The lack of definition of the system and the attendant poor state of knowledge (high uncertainty) of the health and safety threats (risk) involved make it difficult to be confident that the potential problems could be understood. The result is a penalty to those concepts on paper as opposed to those that have evolved to definitive designs. Of course, those systems that have operating experience and a good safety record have an even greater advantage.

What is generally involved with the various waste reduction concepts under consideration is a trade-off of greater short-term risk (for example, less than 100 years) to achieve a reduction of long-term risk (for example, in the thousands of years). There is great uncertainty about long-term risk even though there is considerable evidence that the health consequences are small. There are considerable uncertainties about the exact magnitude of that small consequence. There is difficulty in having confidence about intervening events thousands of years in the future, and therefore there is high uncertainty. On the other hand, much more is known about the operations involved in the short term resulting in greater confidence in the risk results.

Taking a purely state-of-knowledge approach to risk and considering each phase of operation as mutually exclusive, the following qualitative risk rankings are offered within each phase in order of increasing risk:

- For transmutation, the lowest to highest risk alternatives appear to be

- —light-water reactors,
- —fast reactors, and
- —accelerators.

- For separations, the lowest to highest risk alternatives appear to be

- —PUREX,
- —pyrometallurgy,
- —TRUEX, and

- —on-line processing of liquid fuels.
- For disposal, the lowest to highest risk alternatives appear to be
- —geologic repository and
- —surface.

The primary basis for the above qualitative rankings is that risk analyses have been performed on such operations as nuclear reactors; aqueous reprocessing, and, to some extent, geologic repositories. On the other hand, potential safety concerns have been identified, and little or no definitive risk assessment work has been performed on accelerator-based systems, non-PUREX separation methods, and long-term surface storage facilities.

To the extent that each of the various proposed approaches makes use of some mix of these technologies, the safety risk incurred must reflect the uncertainties associated with the individual technologies.

One of the arguments presented by the proponents of S&T technologies is that they have the potential to reduce the radiological risks of the once-through cycle by making more effective use of fissionable resources to make energy. However, published studies indicate that a major source of radiological risk to the public in the once-through cycle is due to releases generated by mining and milling operations. This suggests that spending money on transmutation and separation may not provide the most effective allocation of resources for reducing radiological risk to the public. There is a strong case for concentrating on the front end of the cycle now, while supporting advanced reactor technology research. This would leave open options to take maximum advantage of evolving technologies in order to better use spent fuel as an energy resource in the future.

Risk And Safety Summaries: Radioactive Waste Treatment Concepts

Concept: Accelerator Transmutation of Radioactive Waste (ATW)

1. Transmutation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- A detailed conceptual design of an ATW concept does not exist.
- Many elements of the concept have not been demonstrated.
- (2) *Quantitative Safety Assessments*

- No significant and quantitative risk assessment exists for the ATW,

(3) *Experience Demonstrating Feasibility*

- Design is still in the preliminary concept phase.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Designed specifically to eliminate both actinides and long-lived fission products.

(2) *Unique Risk-Reduction Features*

- Operates below criticality and has rapid system response times.
- Continuous processing of liquid fuel reduces source term and decay heat load.
- Fuel can be drained to subcritical holding facilities with decay heat removal.

(3) *Unique Safety Concerns*

- Must demonstrate ability to maintain system integrity at the beam/ target/blanket interface.
- Liquid fuel eliminates solid lattice and cladding barriers to releases.
- High, localized beam power can initiate a variety of events.
- The high power density in the blanket (150% greater than in a PWR) creates a shutdown cooling problem due to decay heat. Processing rates probably will not be sufficiently rapid to mitigate the most severe shutdown heating transient with loss of normal cooling.
- Large reactivity swings are expected in going from cold shutdown to hot operating conditions.
- ATW concepts appear to be vulnerable to xenon oscillations.
- The nonaqueous ATW fueled partly or completely with thorium will be subject to a reactivity transient safety issue at high neutron flux.

(4) *Uncertainties*

- Beam power of an ATW system would be on the order of 100 times greater than the Los Alamos Meson Physics Facility.
- Behavior of liquid fuel in blanket and during transport through system is not well known.
- Nonequilibrium operations not yet addressed.

2. Separation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- No design for on-line processing of molten salt.
- Requires preprocessing of fuel.
- More extensive research and development necessary than even that required for the ALMR concepts.

(2) *Quantitative Safety Assessments*

- No detailed concept evaluation of either the individual systems or their potential interactions.

(3) *Experience Demonstrating Feasibility*

- None.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- Aimed primarily at destruction of actinides and long-lived fission products.

(2) *Unique Risk-Reduction Features*

- Reduced source term and decay-heat load as a result of on-line fission-product removal.
- (3) *Unique Safety Concerns*

- Multiple operations required to continuously process fuel.
- Short delay times for liquid processing result in higher specific activities during processing. Leads to higher heat generation rates as well.

(4) *Uncertainties*

- Separation methods yet to be defined.
- Chemical stability and system interactions during transients.

3. Storage Phase

a. Status of Evidence

- Same body of information applies to storage of waste from all concepts.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Primary motivation for accelerator-based transmutation is to eliminate the requirements for geologic storage.

(2) *Unique Risk-Reduction Features*

- Concept advocates a mix of facilities that would minimize geologic storage requirements.

(3) *Unique Safety Concerns*

- Activation of replaceable components such as beam targets may increase short-term storage requirements.

Concept: Accelerator (Phoenix)

1. Transmutation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- A detailed conceptual design of the Phoenix concept does not exist.
- Many elements of the concept have not been demonstrated.

(2) *Quantitative Safety Assessments*

- No detailed risk and safety analysis has been performed.
- Almost no analysis of target performance has been made.

(3) *Experience Demonstrating Feasibility*

- Design is still in the preliminary concept phase.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Designed specifically to eliminate actinides and long-lived fission products.

(2) *Unique Risk-Reduction Features*

- Operates below criticality.
- Conceptually simple as a result of direct beam impingement on minor actinides (MA) target.

(3) *Unique Safety Concerns*

- Decay heat is a concern even with a rapid removal of the proton source.
- Reactivity scenarios will have to be addressed.
- Behavior of MA target under direct irradiation by protons must be evaluated.

(4) *Uncertainties*

- A very preliminary concept.
- Performance of target under combined proton and neutron irradiation unknown.

2. Separation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- Proposes conventional aqueous separation with the CURE system.
- Can build upon established processing technology (e.g., PUREX).

(2) *Quantitative Safety Assessments*

- No detailed concept evaluation of either the individual systems or their potential interactions.

(3) *Experience Demonstrating Feasibility*

- None.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Envisions postprocessing to low-level waste.

(2) *Unique Risk-Reduction Features*

- Fuel performance similar to the experience of the FFTF.

(3) *Unique Safety Concerns*

- Multiple processing (spent-fuel and target-fuel) schemes not developed.

(4) *Uncertainties*

- Postirradiation processing uncertain, since isotopic make-up different from fission reactor fuels.

3. Storage Phase

a. Status of Evidence

- Same body of information applies to storage waste from all concepts.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Primary motivation for accelerator-based transmutation is to eliminate the requirements for geologic storage.

(2) *Unique Risk-Reduction Features*

- Concept advocates a mix of facilities that would minimize geologic storage requirements.

(3) *Unique Safety Concerns*

- Activation of replaceable components may increase short-term storage requirements.

Concept: Advanced Liquid Metal Reactor (ALMR)/Integral Fast Reactor (IFR)

1. Transmutation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- IFR concept demonstrated by the EBR-II, but commercial scale plant experience limited.
- ALMR in design phase.

(2) *Quantitative Safety Assessments*

- EBR-II PRA produced core damage estimates an order of magnitude less than core damage frequencies developed in NUREG-1150 for LWRs. EBR proponent states that the EBR-II PRA provides a comprehensive database for the safety risk assessment of the IFR concept.
- PRA of the ALMR, published as GEFR-00873 in 1989, resulted in a frequency of severe core damage of less than 10^{-6} per year. No uncertainty analyses were presented with the results provided.
- (3) *Experience Demonstrating Feasibility*
- The Fast Flux Test Facility has demonstrated high reliability and good performance characteristics.
- While EBR-II and FFTF experience have been favorable, there have been difficulties with some fast reactors. Specific examples are fuel damage in the Fermi reactor and the EBR-1, cracking of primary system components in a U.K. fast reactor, and steam generator leaks in France's Super Phénix reactor and the former Soviet Union's BN-350.
- EBR-II has operated since 1963, producing 60 MWe.
- A significant number of liquid-metal-cooled reactors have been operated worldwide. However, most are either experimental or one-of-a-kind.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Implementing the schedule proposed by the Department of Energy (DOE) could result in no need to load the first repository for high-level waste with spent nuclear fuel.

(2) *Unique Risk-Reduction Features*

- Liquid-metal coolant with large heat capacity and inherent mechanisms to mitigate accidents.

- Capability of safe passive shutdown demonstrated on EBR-II.

(3) *Unique Safety Concerns*

- The ALMR risk model has not been tested against plant-and site-specific considerations found to be important in LWRs.
- Need investigation of dependent failures involving support systems and human response and detailed consideration of long-term accident-recovery capability.

(4) *Uncertainties*

- Little experience with liquid-metal fast reactors on a commercial scale.

2. Separation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- A second, as yet undemonstrated, separation process is necessary for periodic recycling of the ALMR fuel. Pyroprocessing pilot plant studies have been initiated.
- An undemonstrated separation methodology, either aqueous or pyroprocessing, is required to extract transuranics from LWR spent fuel to burn in ALMR.

(2) *Quantitative Safety Assessments*

- A full-scale PRA for a commercial-scale IFR fuel-cycle facility has not been performed.
- Final safety analysis report for the refurbishment of the EBR-II fuel-cycle facility has been prepared and is being reviewed by DOE.

(3) *Experience Demonstrating Feasibility*

- The original EBR-II fuel-cycle facility operated from 1964 to 1969 and was based on early pyrometallurgical processes.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- To reduce the long-term waste storage to require only that fission products be separated from low-level products.

- Use depleted uranium as a feed source, thus reducing the radiological risk from the mining and milling processes.

(2) *Unique Risk-Reduction Features*

- Processing delayed until the iodine and xenon source terms have decayed, thus greatly reducing major contributors to thyroid dose and external dose.
- Processing not to involve high energy sources.
- Batch-oriented processing enhances criticality safety.

(3) *Unique Safety Concerns*

- The risks associated with the untransmuted actinides in the aboveground fuel cycle.
- Pyrometallurgical processes not demonstrated on a large production scale. Pilot-plant work is in progress.

(4) *Uncertainties*

- Lack of experience on a commercial scale.

3. Storage Phase

a. Status of Evidence

- Same body of information applies to storage waste from all concepts.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Geologic repository still required, but ALMR/IFR is one of several means of deferring the need for a second repository.

(2) *Unique Risk-Reduction Features*

- No unique features identified.

(3) *Unique Safety Concerns*

- No unique safety concerns identified.

(4) *Uncertainties*

- Impact of multiple recycling on decontamination factor effectiveness.

Concept: Light-Water Reactors (LWRs)

1. Transmutation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- Little experience with recycled MA fuels; considerable experience with uranium and plutonium.
- Design changes are evolutionary.
- Only considered as potential transmutors in the CURE concept, and role is uncertain.

(2) *Quantitative Safety Assessments*

- No extensive quantitative safety studies of LWRs operating with actinide fuels.
- Many PRAs of existing LWR nuclear plants have highlighted vulnerabilities to improve in next-generation reactors.
- Probabilistic risk assessments of ALWR designs indicate substantial reductions in risk, perhaps of an order of magnitude.
- Next-generation designs in various stages of assessment as part of their certification process.

(3) *Experience Demonstrating Feasibility*

- Considerable commercial operating experience with existing designs that use slightly enriched uranium.
- Minimum operating experience with actinide fuels.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Fissioning reduces actinide inventory.
- Target/blanket designed to transmute long-lived fission products.

(2) *Unique Risk-Reduction Features*

- Performance with low-enriched fuels well established through actual operating experience.

(3) *Unique Safety Concerns*

- Larger concentration of transuranics in the fuel than in once-through cycles requires verification of reactivity control.
- Safety implications of actinide-containing nuclear fuels need to be established.
- Active systems are required to successfully mitigate accidents.

(4) *Uncertainties*

- Commercial experience has not involved actinide-containing fuels.
- Predominant commercial experience limited to slightly enriched uranium.

2. Separation Phase (PUREX process)

a. Status of Evidence

(1) *Systems Technology Maturity*

- Reprocessing of oxide fuels well established.

(2) *Quantitative Safety Assessments*

- Risk assessments available for PUREX proces.

(3) *Experience Demonstrating Feasibility*

- Good experience base for standard reprocessing.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Creates separate actinide and fission product streams.

(2) *Unique Risk-Reduction Features*

- High state of knowledge of separation processes.

(3) *Unique Safety Concerns*

- Disposition of the nonfissile actinides.
- Spontaneous fissioning of some actinides increases radiological hazards during reprocessing.

(4) *Uncertainties*

- Long-term operation of closed fuel cycle.

3. Storage Phase

a. Status of Evidence

- Same body of information applies to storage waste from all concepts.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Reduction of storage requirements only as part of a broader concept.

(2) *Unique Risk-Reduction Features*

- Thermal neutron spectrum is the most effective transmutor for many fission products, thus reducing storage requirements for some long-lived isotopes.

(3) *Unique Safety Concerns*

- None identified.

Concept: PBR

1. Transmutation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- Concept feasibility not yet demonstrated.

(2) *Quantitative Safety Assessments*

- Safety analysis not yet undertaken.

(3) *Experience Demonstrating Feasibility*

- No experience at the power densities (5 MW/liter) and gas flow rates (275 m/s) envisioned.
- Brookhaven National Laboratory believes that many key technologies have been demonstrated in the military and HTGR experience.

b. Selected Design Features

(1) *Applicability to Waste Objectives*

- Designed to burn plutonium, MAs, and long-lived fission products.

(2) *Unique Risk-Reduction Features*

- Low inventories of plutonium, MA, and fission products in reactor.

(3) *Unique Safety Concerns*

- The effects of burn-up reactivity swings on the operations of such a reactor remain to be evaluated, together with the power distribution issues characteristic of high-thermal-flux systems.
- Very high power density (5 MW/liter) creates a safety concern.
- The very high gas-coolant velocity (275 m/s) must be evaluated in terms of emergency cooling requirements.
- Agglomeration of larger particles could lead to hot spots and containment problems.

(4) *Uncertainties*

- Entire reactor concept is very preliminary.
- 2. Separation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- Technology not demonstrated on a total system level.
- Technology supported by HTGRs and weapons-related reactors.

(2) *Quantitative Safety Assessments*

- No published integrated system safety studies available on either the reactor or the proposed processing facilities.
- (3) *Experience Demonstrating Feasibility*
- Reprocessing of PBR fuel not yet demonstrated.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- Burn plutonium, MAs, and long-lived fission products.
- Store cesium and strontium in an monitored retrievable storage, recycle uranium in LWRs, and recycle unburned plutonium into LWRs as MOX fuel.
- Dispose of short-lived fission products as low-level waste after decay.
- (2) *Unique Risk-Reduction Features*
- Long-term risk greatly reduced at the expense of greater short-term risk associated with operations.
- (3) *Unique Safety Concerns*
- Safety of reprocessing PBR fuel is not understood.
- (4) *Uncertainties*
- Highly dependent on processes not yet demonstrated (TRUEX, fluoride volatility, electrolysis, etc.).
- 3. Storage Phase

a. Status of Evidence

- Same body of information applies to storage waste from all concepts.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- The PBR aims to eliminate the need for geologic disposal of high-level wastes.
- (2) *Unique Risk-Reduction Features*

- Thermal neutron spectrum is the most effective transmutor for many fission products, thus reducing storage requirements for some long-lived isotopes.
- (3) *Unique Safety Concerns*
- None identified.
- (4) *Uncertainties*
- Ability to eliminate geologic disposal depends on transmutation and separation efficiencies.

Concept: Clean Use of Reactor Energy (CURE)

1. Transmutation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- Considers both ALMRs and LWRs, but its emphasis is on ALMRs.
- Risks are dependent on mix chosen and evaluation of individual systems.
- Integrated system concepts in early stage of development.
- (2) *Quantitative Safety Assessments*
- Estimated risk will be dependent on evaluation of individual reactor types.
- (3) *Experience Demonstrating Feasibility*
- See experience for individual reactor types.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- Mix of fast and thermal reactors can reduce both actinides and long-lived fission products.
- (2) *Unique Risk-Reduction Features*
- See advantages for individual reactor types.

- (3) *Unique Safety Concerns*
- See concerns for individual reactor types.
- (4) *Uncertainties*
- CURE is simply a concept for mixing other proposed technologies.
- Uncertainties are dependent on both the choice of technology and the mix.
- Largest uncertainty is a method for transmutation of actinides in the near term.
- 2. Separation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- Advocates aqueous reprocessing and separations technologies.
- Well adapted to continuous operation and simple process control.
- The stage for separating cesium, strontium, and targeted isotopes for storage and geologic disposal requires significant technology development.
- (2) *Quantitative Safety Assessment*
- No published integrated system safety assessments.
- (3) *Experience Demonstrating Feasibility*
- Considerable experience with several stages of the CURE concept.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- Mix of fast and thermal reactors with modern aqueous reprocessing technology forms basis for a long-range development program to minimize waste.
- (2) *Unique Risk-Reduction Features*
- Proponents claim CURE with transmutation substantially reduces repository risks for both intrusive and leakage scenarios.
- (3) *Unique Safety Concerns*

- The risk of multifaceted operations, including some not yet developed, still needs to be examined.
- (4) *Uncertainties*

- Little is known about the step to recover the residual transuranics plus technetium.
- A large uncertainty with the CURE-type philosophy is a method for transmutation of the MAs in the near term.
- 3. Storage Phase

a. **Status of Evidence**

- Same body of information applies to storage waste from all concepts.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- CURE aims to make both the liquid reprocessing stream and the solid waste stream qualify for a low-level waste category.
- (2) *Unique Risk-Reduction Features*

- Concept advocates a mix of facilities that would minimize geologic storage requirements.
- (3) *Unique Safety Concerns*

- None identified.
- (4) *Uncertainties*

- None uniquely applicable to CURE.

Concept: Base Case (civilian/military)

1. Transmutation Phase

a. **Status of Evidence**

(1) *Systems Technology Maturity*

- No transmutation.
- (2) *Quantitative Safety Assessments*
- No transmutation.
- (3) *Experience Demonstrating Feasibility*
- No transmutation.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- No transmutation.
- (2) *Unique Risk-Reduction Features*
- No transmutation.
- (3) *Unique Safety Concerns*
- No transmutation.
- (4) *Uncertainties*
- No transmutation.
- 2. Separation Phase

a. Status of Evidence

(1) *Systems Technology Maturity*

- No back-end separation required.
- (2) *Quantitative Safety Assessments*
- No back-end separation required.
- (3) *Experience Demonstrating Feasibility*

- No back-end separation required.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- Permanent storage of unprocessed waste.
- (2) *Unique Risk-Reduction Features*
- Once-through fuel cycle eliminates requirement for separation processes involving high-level waste.
- (3) *Unique Safety Concerns*
- Requires continued mining, milling, conversion, and enrichment, which is a contributor to the radiological hazards of nuclear power.
- 3. Storage Phase

a. Status of Evidence

- Same body of information applies to storage waste from all concepts.
- **b. Selected Design Features**

(1) *Applicability to Waste Objectives*

- All actinides and fission products remain to decay naturally.
- (2) *Unique Risk-Reduction Features*
- None identified
- (3) *Unique Safety Concerns*
- Magnitude of geologic storage requirements is a prime motivation for this evaluation.
- (4) *Uncertainties*
- Uncertainty regarding geological stability and isotopic transport is a prime motivation for this evaluation.

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Appendix J

Fuel Reprocessing Economics

Introduction

Transmutation of transuranics (TRUs) and fission products that are recovered from spent fuel offers potential for improving the technology for long-term disposal of radioactive waste. Successful implementation of this integrated reprocessing/irradiation strategy would require a major financial commitment to the development, design, construction, and operation of a series of reprocessing plants and transmutation reactors based largely on as-yet unproven technology.

This appendix addresses various economic issues related to the use of transmutation as a primary waste management strategy, focusing primarily on future reprocessing costs under various plant ownership and financing arrangements. It also addresses the substantial institutional barriers that would inhibit private-sector financing of such a strategy in the United States.

Historical Perspective

The nuclear industry of the early 1970s was characterized by rapidly increasing worldwide demand for generating commercial nuclear power capacity, rising uranium prices, and an impending shortage of uranium enrichment capacity. There was a consensus within the nuclear community that spent reactor fuel would be reprocessed to recover residual uranium and plutonium for recycle in light-water reactors (LWRs) and ultimately to fuel breeder reactors. Projected reprocessing costs were low, and there appeared to be an urgent need to reduce the rapidly increasing demand for virgin uranium in order to stem rising prices and prepare for early introduction of breeders. U.S. enrichment capacity was sufficiently committed by 1972 that the U.S. Atomic Energy Commission (AEC) closed the books on new orders for enrichment services.

Reprocessing and uranium and plutonium recycle in LWRs was expected to reduce total fuel-cycle cost relative to a once-through fuel-management scheme. Confidence that a reduction in cost would be realized was supported by commercial contracts for reprocessing services at unit prices below $20/kgHM in the late 1960s. The initial estimates of the cost premium for MOX fuel fabrication were as little as 20% above the comparable cost of fabricating virgin uranium fuel. Spent fuel was expected to be reprocessed within approximately 6 months following discharge from the reactor, so that recovered uranium and plutonium could be returned to the reactor with minimal delay.

Through the late 1960s, the U.S. outlook for nuclear power plants and for fuel-cycle facilities to undertake commercial uranium and plutonium recycle facilities was favorable. Two commercial facilities for reprocessing LWR spent fuel were constructed. The General Electric plant at Morris, Illinois, was completed in 1968, and the Allied-General Nuclear Services plant at Barnwell, South Carolina, was completed in 1974. However, neither plant went into commercial service.

The 20-year period of nuclear optimism began to wane in about 1970 with growing public apprehension over the possibility of a major nuclear accident. This in turn prompted criticism of the AEC, which had the dual responsibilities for developing and promoting nuclear power and for regulating its implementation in the private sector. The passage in 1970 of the National Environmental Policy Act (NEPA) fundamentally changed the ground rules for nuclear plant siting and construction. NEPA added a requirement for the preparation of an environmental impact statement for all new major facilities, with provisions for public hearings on the statement before a construction permit could be issued.

Public dissatisfaction with AEC management of nuclear issues heightened with the revelations attending the cancellation of the planned high-level waste (HLW) repository near Lyons, Kansas. The public became aware that an approved waste repository site had not yet been secured to receive the steadily increasing quantity of spent fuel, as well as HLW from defense operations. In 1974 the Ford administration divided the AEC into two new organizations that separated the responsibilities for nuclear development and regulation. The new organizations were the Energy Research and Development Administration (ERDA) and the Nuclear Regulatory Commission (NRC). At the same time, Congress disbanded the powerful Joint Committee on Atomic Energy and reassigned its responsibilities among several congressional committees.

A new public concern arose when India tested a "peaceful nuclear explosion" in 1974, rekindling an earlier controversy over the postulated link between nuclear power and nuclear weapons proliferation. This concern was heightened by the revelation that the Indian nuclear explosive contained nuclear weapons material produced in a reactor that was sold to India by Canada. The Indian nuclear test made it necessary for many nations, including several developing countries, to acquire their own facilities to enrich uranium or recover plutonium from spent reactor fuel. The growing controversy associated with the widespread commercial use of plutonium recycle came to a head in the highly contentious hearings on the Generic Environmental Statement on Mixed Oxide (GESMO) fuel, which the NRC had begun in late 1974 and which the Carter administration finally canceled in 1977.

In 1977, the Carter administration completed a review, begun by the Ford administration, of the plans to commercialize the breeder reactor and engage in plutonium recycle in the United States. In addition to placing greater emphasis on nonproliferation concerns, this review was based on projections of the growth in electric power demand that were much lower than the earlier projections of the AEC up to the mid-1970s. This reappraisal soon led to cancellation of breeder commercialization and plutonium recycle, leaving the Barnwell reprocessing plant without an operating license or a mission.

From 1977 to 1979, the United States joined other nations in the International Fuel Cycle Evaluation to reconsider the commercial use of plutonium. The U.S. Congress passed the Nuclear Nonproliferation Treaty of 1978, which placed restraints on foreign reprocessing of nuclear fuel of U.S. origin.

Several nations took issue with the change in nuclear ground rules implemented unilaterally by the United States. However, most ultimately agreed that steps to limit reprocessing and recycle would be prudent, although they retained reprocessing as a long-term option. The International Fuel Cycle Evaluation also reached agreement that spent fuel itself was a waste form that could be safely disposed of in a waste repository. Indeed, by 1977, the United States was strongly recommending that course, which was adopted and further promoted by several other nations, notably Sweden, that had small nuclear programs. However, the European reprocessors and Japan continued their plans to reprocess spent fuel from commercial reactors and to store the separated plutonium pending further development of their breeder programs.

When the U.S. ban on domestic fuel reprocessing was lifted in the early 1980s, the previously favorable political and institutional environment for nuclear energy no longer existed, and the economic incentives for reprocessing had changed markedly. The principal factors precluding the resumption of commercial reprocessing in the United States were as follows:

- There was widespread cancellation of orders for new U.S. nuclear power plants, resulting in a precipitous decline in uranium concentrate prices from a peak of \$44/lb U_3O_8 in mid-1979 to approximately \$17/lb only 3 years later.
- There was greatly increased worldwide uranium ore reserves as a result of major discoveries in Australia in the late 1970s and in Canada in the early 1980s.
- The introduction of commercial gas centrifuge enrichment technology, completion of the Eurodiff gaseous diffusion plant, and the absence of new orders for U.S. nuclear power plants transformed the earlier forecast of a shortage of enrichment capacity to a condition of oversupply, resulting in major price reductions for enrichment services due to intense competition.
- The Nuclear Waste Policy Act of 1982 provided no economic incentive for HLW vitrifications as opposed to direct disposal of spent fuel. Indeed, the disposal of spent fuel in a geologic repository remained the Department of Energy (DOE) policy, in conformance with the Nuclear Nonproliferation Treaty of 1978.
- The capital cost of breeder reactors relative to LWRs proved to be higher than had been projected in the early 1970s, and the unit cost of fabricating MOX LWR and liquid-metal reactor (LMR) fuel also proved higher than projected, exhibiting a steady increase with time. Large-scale deployment of commercial breeder reactors, once envisioned to begin as early as the 1980s, was delayed indefinitely. It now appears that the need for an LMR to forestall rising uranium costs may not occur until the second half of the next century, or later.
- The estimated cost to construct and operate commercial reprocessing plants increased due to several factors, including new regulations requiring containment of radioactive gases and solidification of the plutonium product, as well as the industry's recognition that fuel reprocessing should be treated as a high-risk venture, thereby increasing plant financing cost.
- There was a lack of a clear economic incentive for reprocessing and closure of the LWR fuel cycle as a result of the above events. Reprocessing and recycle of plutonium would have required reopening of the contentious GESMO hearings.

These formidable barriers to reprocessing led U.S. utilities to continue with a once-through fuel cycle and prompted reoptimization of LWR fuel management schemes to use longer and higher burn-up cycles. This reduced the fissile uranium and plutonium content of the spent fuel, further reducing the incentive to reprocess.

Foreign interest in reprocessing continued, however, as those nations with limited economically recoverable domestic uranium resources saw reprocessing as an opportunity to reduce the increasing demand for uranium imports. In France, the 800-MTHM/yr UP3 reprocessing facility, as well as the 400-MTHM/yr expansion of UP2, were successfully designed, constructed, and operated. Construction of the 900-MTHM/yr Thermal Oxide Reprocessing Plant (THORP) in the United Kingdom was completed in 1992, and the facility is currently in start-up. In Japan, the 800-MTHM/yr Rokkashomura plant is currently under construction and is scheduled for operation in 2000.

Relative Economics of Reprocessing Versus Once-Through Fuel Cycle

A study of LWR fuel-cycle costs was recently performed by the Nuclear Energy Agency (NEA) of the Organization of Economic Cooperation and Development (OECD). Their report, *The Economics of the Nuclear Fuel Cycle* (OECD/NEA, 1993), concluded that the levelized fuel cost for the once-through LWR fuel cycle is approximately 14% less than for the reprocessing cycle. Their analysis included a credit for both the uranium and the plutonium recovered through reprocessing.

Those countries that chose to reprocess their nuclear fuel based their decision on a number of factors other than cost. With limited natural uranium resources, they have a strong interest in being self-sufficient in energy production. They also take a longer-term look than the "what are the profits in the next quarter" attitude of many of the decisions made in the United States. This led them to a strategy that included recovery and recycle of uranium and plutonium from spent fuel and vitrifying the HLW produced as a solid waste form for interim storage until a permanent waste repository is in operation.

Principal Issues in Determining Whether to Adopt Reprocessing and Transmutation as a Waste Management Strategy

Recycling and transmuting TRUs and fission products requires reprocessing. While transmutation proponents claim that this can be done as economically as a once-through fuel cycle, the cost of reprocessing is a key factor in the cost of transmutation. Marshaling the management, technical, and financial resources to design, construct, and operate the reprocessing plants and specialty reactors to recycle and transmute TRUs and fission products is influenced by a number of major issues. While certain issues are largely political/institutional, they are nevertheless important, since they will influence the attitude of prospective lenders for these capital-intensive projects.

The principal issues are

- the relative immaturity of certain aspects of nonaqueous (pyrochemical) reprocessing technology and of the advanced liquid-metal reactor (ALMR) technologies proposed to recycle and transmute the TRUs and fission products, and the magnitude of the development and demonstration program required before wide-scale implementation of a transmutation strategy can be implemented;
- likely growth in the required capital, operating and maintenance, and decommissioning costs for pyrochemical reprocessing and ALMR power plants as concept definition evolves;
- difficulty in obtaining a government financial commitment because of the expected high cost of transmutation technology development/implementation and the difficult-to-quantify benefits to public health and safety; and
- difficulty in attracting private capital due to the perceived high technical/economical/institutional risk of reprocessing/transmutation projects relative to alternative opportunities for investment capital, resulting a higher cost of capital due to the higher perceived risk.

These issues apply not only to centralized, large-scale reprocessing plants but also to smaller onsite reprocessing plants, transmutation reactors, and an integrated complex of reprocessing plants and transmutation reactors. Overcoming these barriers will be a formidable challenge.

Reprocessing-Plant Capital Costs

While pyroprocessing technology is being considered for reprocessing spent fuel that is associated with transmutation of TRUs, this technology is not sufficiently mature that reliable reprocessing-plant capital and operating cost estimates can be prepared. Moreover, it is by no means certain that pyroprocessing will prove more economical than conventional aqueous reprocessing, for which the technology is relatively mature. While Argonne National Laboratory (ANL) prefers pyroprocessing technology, the fuel-cycle costs developed by General Electric (GE) for the ALMR program are apparently based on the cost of plutonium and uranium recovery by extraction/transuranic extraction (PUREX/TRUEX) aqueous technology for reprocessing LWR spent fuel to start and refuel ALMRs. The GE estimates are based on PUREX aqueous technology, combined with new TRUEX technology for high-yield recovery of all TRUs. The capital costs developed in this section, and the operating costs developed in the section *Reprocessing-Plant Operating Costs* are therefore based on aqueous reprocessing technology.

Capital Costs Associated With Reprocessing Plant Projects

Capital cost information was obtained from the open literature and private communications for three reprocessing plant projects: THORP (United Kingdom), UP3 (France), and Rokkashomura (Japan). UP3 is in operation, THORP is in the startup phase, and Rokkashomura is under construction. These three plants have annual throughputs in the range of 800 to 900 MTHM/yr, and each is based on aqueous PUREX technology.

Thermal Oxide Reprocessing Plant (Thorp)

The THORP reprocessing plant, which is located at the Sellafield site in the United Kingdom, is owned by British Nuclear Fuels, plc. THORP services include fuel receipt and interim storage; reprocessing (including conversion of uranium and plutonium to oxide); HLW vitrification and intermediate storage; intermediate-level waste encapsulation, interim storage, and disposal; and low-level waste (LLW) disposal. The reprocessing-plant capital and operating costs used in this appendix are based on this scope of services.

The reported capital cost of the THORP facility ranges from £2,600 million ($4,700 million in 1992 dollars) reported by Wilkinson (1987) to £4,000 million ($6,800 million) reported by K. Uematsu (private communication, 1992). The most credible cost is £2,850 million ($4,560 million), which was reported in the July 1993 report *The Economic and Commercial Justification for THORP* (British Nuclear Fuel, 1993). This cost excludes interest during construction since the plant was financed through up-front payments by THORP customers.

The average annual plant throughout scheduled for the initial 10 years of operation is 700 MTHM/yr, but British Nuclear Fuel estimates that the plant can process 900 MTHM/yr (1,200-MTHM/yr capacity, with operation at a 75% capacity factor). The OECD/NEA study, which based its reprocessing plant costs on the THORP facility, assumed a plant throughput of 900 MTHM/yr as well.

UP3

UP3 is the most recent addition to the large French reprocessing complex at La Hague. It reportedly has a design capacity (annual throughout) of 800 MTHM/yr. UP3 has been in operation since 1990, providing complete services that range from spent-fuel storage through HLW vitrification.

COGEMA, the owner/operator of UP3, reported a total capital cost of *ff* 50 billion for UP3 plus the 400-MTHM/yr expansion of UP2 to 800 MTHM/yr. If two-thirds of the cost is allocated to UP3 (based on the ratio of plant capacities), the resulting UP3 capital cost would be approximately $5,800 million. COGEMA also reported that the design and construction of UP3 required 25 million engineering man-hours and 56 million man-hours of field construction (*Reprocessing News*, 1990). While no information was provided on the cost of equipment, materials, interest during construction, and other such costs, factoring in these costs based on historical experience results in a capital cost of $6,670 million to construct a comparable facility in the United States.

Rokkashomura Plant

Japan Nuclear Fuel, Ltd. is currently constructing an 800-MTHM/yr reprocessing plant at the Rokkasho Village site. It is scheduled for operation in 2000. The technology is primarily French, with U.K. and German design input as well. This facility provides complete services, including HLW vitrification and storage for 8,200 waste canisters. The products are uranium and plutonium, with the plutonium diluted with uranium and converted to a 50/50 mixture of uranium/plutonium oxide.

Despite the lessons learned on the design and construction of the UP3 and THORP reprocessing plants, the constant dollar capital cost at Rokkashomura has not decreased significantly relative to these earlier plants. Reported capital costs for this facility range from $6,500 million (Chang, 1992) to $5,200 million (¥740 billion) (JGC, 1991). Because of the particularly stringent seismic design requirements in Japan, the cost of a facility of comparable capacity would probably be less in a region of lower seismicity.

Capital Costs Associated With Reprocessing Plant Studies

Capital costs associated with a number of studies were also evaluated, the most important being the June 1993 "final revised draft" of the OECD/NEA study *The Economics of the Nuclear Fuel Cycle* (OECD/NEA, 1993). The committee considers this study particularly credible, because it is based on cost data from the recently completed THORP plant and includes input from COGEMA, the owner/operator of UP3. The capital costs reported in the OECD/NEA study are shown in Table J-1.

The total estimated cost of £3,297 ($5,370) million is 15% higher than the reported actual cost of THORP of £2,850 million ($4,560 million without interest during construction). The reason for this cost difference is not evident, but it could be associated with the higher cost of construction at the "grass roots" site assumed in the OECD/NEA study as compared with the existing site on which THORP is located.

In addition to the above capital-cost estimates for actual plants and the OECD/NEA study, a number of other studies of reprocessing plant economics have presented estimates over the past decade. Examples include the following:

- The 1990 study for a generic U.S. site estimated reprocessing-plant capital costs ranging from $2,725 million (government-owned plant) to $3,001 million (privately owned plant) for a reprocessing plant with an annual throughput of 1,500 MTHM/yr, 67% larger than the 900-MTHM/yr throughput of THORP (Gingold et al., 1991). These costs assumed a mature

TABLE J-1 OECD/NEA Reprocessing-Plant Capital Cost Estimate (900 MTHM/yr annual throughput).

Cost Component	Capital Cost[a] (1991 £ millions)
Fuel Receipt and Storage	100
Reprocessing Plant	2,300
High-Level Waste	
Vitrification	260
Interim storage	59
Intermediate-Level Waste	
Encapsulation	300
Interim storage	38
Site Preparation and Services	
Site preparation	229
Site services	11
Total Capital Cost	
1991 £ millions	3,297
1992 $ millions	5,370

- [a] Excludes interest during construction (see Table J-3).

industry. The reprocessing plant involved aqueous PUREX technology together with new TRUEX technology for high-yield recovery of all TRUs.

- The ALMR fuel-cycle assessment (M.L. Thompson, private communication, 1990) developed a capital cost of $6,100 million for a 2,700-MTHM/yr reprocessing plant, three times the capacity of THORP. This cost also includes facilities for MOX fuel fabrication services. As in the Gingold et al. study, the reprocessing plant involved aqueous PUREX technology together with new TRUEX technology.
- McDonald (1993) presented a capital cost of $4,380 million for THORP (escalated from as-spent dollars to 1992 dollars).
- An OECD/NEA report (1989), derived unit reprocessing costs based on an analysis of the THORP plant. Reported reprocessing costs were $570/kgHM for a 5%/yr return on capital and $750/kgHM for a 10%/yr return. Depending on the assumptions used for plant economic lifetime, these figures would indicate a plant capital cost ranging from approximately $6,000 to $7,000 million (for a 15 to 30-year plant economic life).

The capital costs estimated in the first two of the above studies are substantially below those experienced in the construction and operation of actual plants. This is particularly difficult to rationalize considering that the study plants include new TRUEX separations and have annual throughputs two to three times higher than THORP, UP3, and Rokkashomura. Assuming that reprocessing plant capital costs are proportional to the 0.6 power of plant capacity, scaling the Gingold estimates to a 900-MTHM/yr capacity would result in a capital cost of $2,010 million for a government-owned facility and $2,210 million for a privately owned facility. Similarly, the GEl capital cost would decrease to $3,160 million for a 900-MTHM/yr plant, less the cost of the fuel fabrication facilities included in the GE estimate. These costs are only one-third to one-half of the costs reported for actual plants of 800 to 900–MTHM/yr capacity.

Even lower estimates of capital costs were presented earlier by the ALMR project (Salerno et al., 1989) for PUREX/TRUEX reprocessing plants designed for high-recovery yield of all TRUs from LWR spent fuel. Two cost estimates were presented, one with an annual throughput of 300 MTHM and a second with an annual throughput of 2,500 MTHM. Their estimates, escalated from 1989 to 1992 dollars, were $227 million and $4,250 million, respectively. Scaling the 300-MTHM/yr plant estimate to 900 MTHM/yr would result in a cost of $440 million, while scaling the 2,700-MTHM/yr plant estimate to 900 MTHM/yr would result in a cost of $2,200 million. These estimates are far below the reported capital costs of actual plants. More important, they indicate an inverse economy of scale with plant throughput, which has not been observed or predicted in other studies.

Based on the above information, the committee concludes that reported capital costs for actual contemporary plants currently provide the most reliable basis for estimating the cost of future plants. Estimated capital costs reported in recent U.S. studies appear inexplicably low.

Summary of Estimates of Capital Cost

Table J-2 summarizes the capital costs reported for the above commercial facilities, as well as costs reported in more recent studies. Costs in year-end 1992 dollars were developed using historical escalation rates in the United States over the period that the plants were under construction. Costs expressed in foreign currencies were converted to U.S. dollars using the average exchange rate over the 10-year period from 1983 to 1992.

Interest During Construction

Since both the THORP and UP3 plants were financed largely through customer prepayments, it is the committee's understanding that the capital costs quoted in Table J-2 for these facilities do not include interest during construction. Financing costs for reprocessing plants are large because of the relatively long construction schedules. UP3 construction required 10 years (1981–1990), and THORP required 10 years (1983–1992). Rokkashomura construction started in 1993 and is scheduled for completion in 2000 (8 years). Preconstruction activities such as siting studies, permitting, and that portion of the engineering performed prior to construction start typically require an additional 2 to 3 years or more.

The OECD/NEA study indicated a construction schedule for the reprocessing plant of 11 years, which the committee assumes represents the total project schedule. Other facilities, such as the spent-fuel storage ponds, waste vitrification facility, and intermediate-level waste storage facility, had shorter schedules ranging from 5 to 8 years. The committee estimated the total cost of interest during construction by summing the financing cost for each of the major facilities.

For the government-owned plant, the interest rate for calculating interest during construction is simply the constant-dollar cost of interest on debt capital (4.0%). For the utility-owned plant, the appropriate rate is the weighted cost of capital (6.4%) (EPRI, 1993). For the privately owned plant, the committee used the cost of debt (8%), assuming project financing. The alternative, financing interest during construction with the average cost of capital (15.3%) appeared to be an unrealistic case due to its extremely high cost.

Since the distribution of the cash flow stream over the project schedule was not known, the committee applied a typical "S" curve to determine cumulative expenditures versus time. The shape of this curve is such that 50% of the constructed cost of each facility will have been spent 65% through the project schedule.

Applying these interest rates to the construction schedule for the various facilities in the reprocessing complex results in the total plant capital costs shown in Table J-3.

TABLE J-2 Reprocessing-Plant Comparison of Capital Cost

			Capital Cost (Billions)		Unit Capital Cost
Reprocessing Plant/Study	Year[a]	Processing Throughput MTHM/yr	Foreign Currency	1992 $	$/kgHM/yr
Reprocessing Plants					
THORP (United Kingdom - Sellafield site)					
Wilkinson (1987)	1987	900	£ 2.6	4.7	5,250
British National Fuel (private Communication, 1989)	1989	900	£ 4.0	6.8	7,500
UP3 (France – LaHague site)					
UP3 + UP2 expansion	1985	800+400	*ff*50	5.8 (UP3)	7,230
UP3 (derived from engineering/construction man-hour estimates)	1992	800	-	7.3	9,100
Rokkashomura (Japan)					
Chang (1990)	1990	800	-	6.5	8,100
Uematsu (private communication, 1992)	1989	800	¥840	5.2	6,510
Reprocessing Studies					
Generic U.S. site (Gingold et al., 1991)	1990	1,500	-	3.4	2,200
Plutonium Fuel - An Assessment (OECD/NEA, 1989)	1989	700	-	6.0–7.0	8,600–10,000
The Economics of the Nuclear Fuel Cycle (OECD/NEA, 1993)	1991	900	£ 3.3	5.4	5,970
Review of Fuel Cycle Costs for the PWR and Fast Reactor (British National Fuel, 1987)	1987	600	£ 1.7	3.1	5,150
ALMR Fuel Cycle Assessment - 1991 (GE, 1991)	1991	2,700	-	6.1	2,300[b, c]

[a] Year is the base year for cost estimate.

[b] Since this facility also provides fuel fabrication services, the unit capital cost for reprocessing only would be somewhat less than $2,300/kgHM of annual plant throughput.

[c] THORP plant capacity changed to 900 MTHM/yr in all cases.

TABLE J-3 Total Capital Cost (1992 $ millions)

Owner	Constructed Cost	Interest During Construction	Capital Cost
Government	5,370	790	6,160
Utility	5,370	1,300	6,670
Private Venture	5,370	1,950	7,320

Capital Cost Uncertainties

Because of commercial considerations, insufficient information is available in the open literature to fully define the scope and financial parameters incorporated in the reported capital costs of these facilities. It is uncertain whether items such as research and development, owner engineering, site acquisition, environmental/regulatory activities, start-up, training, and decommissioning are included. Converting costs expressed in foreign currencies to U.S. dollars introduces considerable uncertainty. The use of customer advanced payments to finance both the THORP and UP3 plants further complicates the analysis of these reported costs. The range of costs reported for such facilities is nevertheless useful to assess the realism of the economic analyses presented in the various studies performed to estimate future LWR and ALMR reprocessing costs.

Foreign Exchange Rates

Converting capital costs expressed in foreign currencies to U.S. dollars introduces considerable uncertainty. As shown in Figure J-1, exchange rates varied considerably over the period during which the THORP and UP3 plants were constructed (*International Financial Statistics* , 1993). Where costs were reported in foreign currencies, the committee used the average exchange rates for the 10-year period from 1983 to 1992:

British pounds	1.60 $/£
French francs	6.70 FF/$
German marks	2.08 DM/$
Japanese yen	170 ¥/$
European Currency Units	1.07 $/ECU

Despite the uncertainties associated with currency conversion, the committee believes that reprocessing-plant capital and operating costs based on or derived from actual plants are superior to those derived from studies, except for the OECD/NEA study (1993) in which costs were derived from the THORP facility.

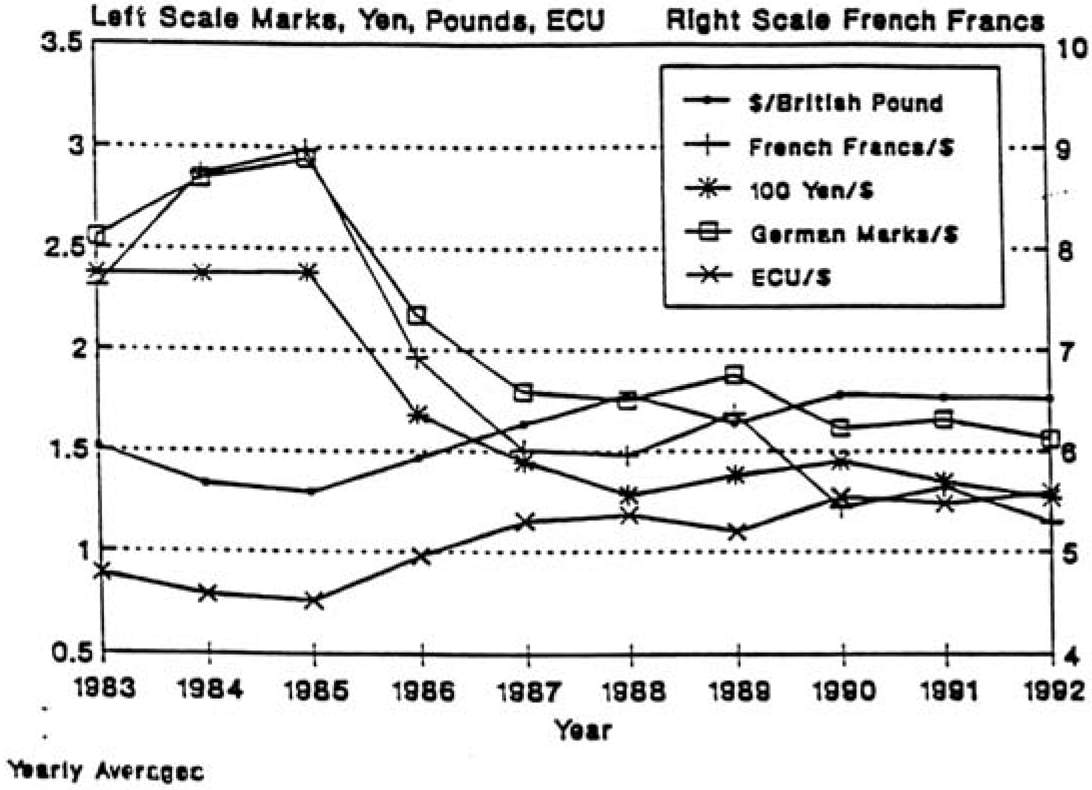

FIGURE J-1 Currency Exchange Rates 1983–1992.

Labor Productivity

Differences in labor productivity between the United States and the United Kingdom, France, and Japan introduce uncertainty in translating foreign capital costs to U.S. dollars. The committee made no attempt to adjust the estimates for such differences.

Start-Up Costs

The committee assumed that the capital cost presented in the OECD/NEA study includes start-up costs. If not, the capital cost would have to be increased substantially (by 50 to 100% of the $374 million annual operating costs).

Reprocessing-Plant Operating Costs

For commercial reasons the amount of definitive information available for reprocessing-plant operating costs is extremely limited.

The OECD/NEA study (1993), reported an annual 1991 operating cost of £230 million ($374 million) based on the THORP plant. As shown in Table J-4, this includes waste processing, interim storage, and disposal (excluding HLW).

TABLE J-4 Operating Cost According to the OECD/NEA Study (900 MTHM/yr annual throughput).

Cost Component	Operating Cost (1991 £ million/yr)
Fuel Receipt and Storage	11
Reprocessing Plant	145
High-Level Waste	
Vitrification	22
Interim storage	2
Intermediate-Level Waste	
Encapsulation	33
Interim storage	1
Disposal	5
Low-Level Waste	
Disposal	11
Total Operating Cost	
1991 £ million/yr	230
1992 $ million/yr	374

British Nuclear Fuel, plc., reported that the present value of avoidable operating costs from not operating THORP for the initial 10 years, discounted at a rate of 2%, would be £1,560 million (British Nuclear Fuel, 1993). This equates to an annual cost of £174 million. Since the £174 million/yr excludes unavoidable costs (magnitude not stated), it would appear to be in reasonable agreement with the OECD/NEA value of £230 million stated above.

Operating costs reported in recent U.S. reprocessing plant studies are substantially below the OECD/NEA and British Nuclear Fuel, Ltd. values. Gingold developed an operating cost range of $179 to $184 million/yr for a 1,500-MTHM/yr reprocessing plant. GE estimated an operating cost of $259-million/yr for their 2,700-MTHM/yr plant. The GE cost appears to include consumables of approximately $145 million/yr associated with fuel fabrication operations. Deleting this cost to reflect reprocessing cost only would reduce operating costs to $114 million/yr, which appears very low in comparison with the estimates for THORP, particularly for a facility with an annual throughput three times larger than THORP.

The committee adopted the OECD/NEA operating cost estimate of £230 million/yr ($374 million/yr), since it is based on THORP and has been subjected to peer review.

Financing of Reprocessing/Transmutation Reactor Facilities

Estimating unit processing costs from the capital and operating costs developed above requires definition of the financial structure of the prospective plant owner/operators and of the associated cost of debt and equity capital. The alternative financial structures might include:

- private venture;
- utility/industry consortium;
- government; or
- hybrid arrangement.

Each of these alternative arrangements is discussed below.

Private Venture

There are clear precedents for private ownership of both electric power generating facilities and chemical process plants. The prime examples of privately owned power plants, other than those owned by electric utilities, are cogeneration plants. These private ventures by independent power producers differ significantly from the proposed fuel-processing/transmutation facilities, however, in that cogeneration projects are based largely on demonstrated technology, and project financing is supported by power sales agreements with the utilities that purchase the electricity generated. The financial risk to the lenders is therefore relatively low.

Typical financing arrangements for cogeneration projects are

- a relatively high debt-to-equity ratio (80–85% debt and 15–20% equity) because of the low perceived risk;
- a nominal 15-18%/yr after-tax return on equity (11–14%/yr in constant dollars);
- a nominal 10-11%/yr cost of long-term debt (6–7%/yr in constant dollars), approximately 300 basis points (3%) above the nominal rate on U.S. long-term treasury bonds; and
- project financing supported by power sales agreements with the utility customer (without these agreements, the return on stockholder equity and the rate of interest on debt financing would have to be substantially higher).

An inflation rate of 4%/yr was assumed to convert nominal rates to constant dollar rates. The rate premium on both equity and debt financing would vary according to the perceived project risk. For example, a cogeneration plant using conventional gas turbines would pose less technical and economic risk than one based on a more advanced technology such as a pressurized fluidized-bed coal-fired boiler.

If it were possible to finance a reprocessing/transmutation project in the private sector, the required after-tax return on equity would be greater than that of current cogeneration projects because of the much higher technical, financial, and political/institutional risks associated with nuclear projects, particularly one based largely on new technology such as pyrochemical reprocessing. The required constant dollar after-tax return on equity might range from 14-18% as compared with 11-14% for a cogeneration project. The proportion of total capital represented by equity would likely be substantially higher than that typically required because of the higher risk associated with transmutation technologies.

With regard to the availability of debt financing, banks typically make loans up to a certain level of risk, with the rate increasing with risk. Beyond that level of risk, however, they are generally not prepared to lend at any rate of interest. The facilities associated with the reprocessing/transmutation complex may fall in this category unless some means of risk reduction (insurance, government risk-sharing, etc.) can be developed.

A pure private venture to design, build, own, and operate such a complex, without government financial guarantees, appears unrealistic. The level of risk appears unacceptably high to obtain the required financing due to:

- immature, unproven technology;[1]
- likely strong opposition from public interest groups;
- high regulatory uncertainty (first-of-a-kind facilities);
- general reluctance of financial community to finance nuclear projects;
- potential adverse future changes in government policy that could preclude plant operation; and
- the Barnwell reprocessing plant experience which is a major deterrent to new private reprocessing ventures.

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[1] This applies to pyroprocessing and new TRU-burning reactors but not to LWR PUREX reprocessing technology, which is considered mature.

The Department of Energy (DOE) assumes that private industry would finance, construct, and operate a large (2,700 MTHM/yr of LWR spent fuel) centralized reprocessing plant to obtain TRUs from spent fuel to start and refuel TRU-burning ALMRs. The plant would have to begin operation in 2010.[2] The new reprocessing plant would be based on presently unproven pyrochemical technology, operating with more than tenfold lower process losses than yet achieved in commercial reprocessing facilities. To justify the market for the TRUs recovered from this new reprocessing plant, the utilities would have to commit to building over 20 large ALMRs. There would be no other market for the TRU product from the reprocessing plant. Being highly contaminated with fission products and minor actinides (neptunium, americium, and curium), the TRUs could not be used as MOX fuel in current LWRs.

To achieve the extent of TRU depletion sought by DOE's ALMR program, many subsequent replacement ALMRs would be required to further transmute the considerable TRU inventory remaining at the end of life of the first-generation ALMRs. Advanced commitments of these subsequent ALMRs and their progeny would also be required to obtain the claimed benefits to waste disposal.

These are hardly the conditions conducive to risk-capital financing of such a venture, particularly of the new and unique plant for reprocessing LWR spent fuel. Federal subsidies, guarantees, or ownership would be required. The extent of commitment that the U.S. government would be required to provide would be enormous and unprecedented.

As far as reprocessing is concerned, the above reference to "immature, unproven technology" refers primarily to pyrochemical rather than PUREX aqueous reprocessing, since the former is in an early stage of development, while the latter is relatively mature. However, even a reprocessing plant using PUREX aqueous technology combined with the newer TRUEX process for high-yield recovery of actinides for transmutation would be unproven and difficult to finance.

Utility-Owned, TRU-Burning Nuclear Power Plant

In general, utilities are conservative, risk-adverse organizations. Their experience with the substantial cost growth resulting from nuclear power plant regulatory delays and adverse changes in regulatory requirements (such as the retrofits resulting from the accident at Three-Mile Island Unit II) has strengthened this management philosophy, making it much more difficult to enlist their participation in high-risk ventures.

The intermediate-term utility industry strategy for selecting future additions to nuclear generating capacity is based primarily on developing advanced LWRs. These evolutionary designs are expected to be founded on proven technology. Utilities understand the risks associated with advanced reactors and are actively participating in setting generic design criteria.

The rationale for utility ownership in a project requiring a special-purpose, non-LWR reactor technology to transmute radionuclides based on first-of-a-kind, nonproven technology

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[2] Operation in 2010 assumes the first ALMR operation in 2012. Some DOE projections show the first ALMR operation as early as 2007, which would require reprocessing to start in 2005.

such as the integral fast reactor (IFR) and the accelerator transmutation of wastes (ATW) concepts is difficult to conceive:

- Since the Nuclear Waste Policy Act of 1982 essentially relieves utilities of the responsibility for postirradiation processing and permanent disposal of spent fuel, they currently have no incentive to get involved as an owner in a high-risk actinide-burning venture.
- Politically sensitive public utility commissions would be unlikely to approve utilities entering into a TRU transmutation venture, particularly when the utilities have no legal requirement or financial incentive to do so. Prudency hearings conducted after substantial financial commitments had been made (or after the plant has been completed) might reverse earlier agreements and disallow previously approved costs.
- At a minimum, it would appear that some new type of government risk/cost sharing, far more extensive than on past projects/programs, would be necessary to attract utility participation.

Government-Owned Facility

Ownership of reprocessing plants in European countries has generally been confined to government corporations. They possess the unique ability to finance projects at low interest rates and are able to undertake complex, high-risk projects in the interest of national policy. Without such low-cost financing (from the government or from advance payments for services from plant-customers, as was the case for the THORP plant), reprocessing costs would be prohibitively high. Since the federal government is ultimately responsible for the safe disposal of spent fuel in the United States, it would be the logical entity to undertake a TRU-burning program, if such were possible within the institutional barriers of government.

Hybrid Organization

Some form of hybrid organization involving government, utilities, and private industry might feasibly own and operate a reprocessing/transmutation complex. Possibilities include government ownership of the reprocessing plants (high risk), with utilities or private investors owning the power plants. Alternatively, the government could own the entire complex, with a private sector operator. The government could also provide special legislation designed to limit project risk (one-stop licensing, long-term contracts for reprocessing and waste transmutation, exemptions from future regulatory changes, etc). For the purpose of this appendix, the committee did not examine the implications of such a hybrid ownership group on project economics.

DOE's ALMR project assumes a different hybrid organization. In this scenario, ALMRs are built and operated by electric utilities and collocated with integral fast reactor (IFR) facilities for receiving and recycling TRUs for ALMR spent fuel. A large, centralized facility for reprocessing LWR spent fuel would be owned and financed by a chemical company as a low-risk venture. Here the institutional barrier would be even higher. Chemical companies normally expect a greater return on investment than do regulated utilities.

Government Ownership—The Only Alternative

Studies by DOE's ALMR project emphasize private ownership and financing of the ALMR reactors and associated reprocessing/fabrication facilities. However, the above discussion of alternative ownership arrangements leads us to the conclusion that government ownership of reprocessing and transmutation facilities is the only viable alternative at the outset. As stated earlier, the government is ultimately responsible for long-term disposal of HLW and is the only institution capable of overcoming the political/institutional issues. This is further supported in subsequent discussions of the relative cost of reprocessing under government, utility, and private (nonutility) financing arrangements.

Hundreds of years of reprocessing/transmutation will be required to achieve the TRU reduction argued by its proponents of transmutation concepts as necessary to permit geologic disposal. The complexity, the high potential costs of the reprocessing and transmutation system, and the extensive period over which the transition system must be committed to operate in order to benefit waste disposal raise serious questions as to the feasibility of such an unprecedented commitment even by the government.

Estimated Unit Costs of LWR Reprocessing

Unit reprocessing costs were calculated for government, utility, and privately owned reprocessing plants, based on the ground rules shown in Table J-5. the assumed return on common equity and interest on debt are expressed in after-tax, constant (inflation-free) dollars. Utility parameters are based on the 1993 *EPRI Technical Assessment Guide* (EPRI, 1993). The cost of plant decommissioning was neglected. The resulting levelized unit reprocessing costs for a capital cost of $7,320 million (includes interest during construction), an annual operating cost of $374 million, and an annual plant throughput of 900 MTHM are as follows:

Plant Owner/Operator	Unit Cost ($/kgHM)
Government	810
Utility	1,330
Private	2,110

Comparison With Published Reprocessing Prices

Reprocessing unit prices reported in the literature for actual plants (THORP and UP3) typically range from approximately $600 to $1,400/kgHM. These prices reflect the relatively

TABLE J-5 Reference Case Parameters

Parameters	Government	Utility	Private Venture
Cost Assumptions			
Capital Cost ($ million)[a]	6,160	6,670	7,320
Operating Cost ($ million/yr)	374	374	374
Property Taxes and insurance (%/yr of initial capital cost)	0	2	2
Annual Refurbishment Cost (%/yr of initial capital cost)	1	1	1
Financial Structure			
Common Stock (%)	0	46	70
Preferred Stock (%)	0	8	0
Debt (%)	100	46	30
Investment Returns and Interest Rates (constant $)			
Common Stock (%/yr)	na	8.5	16.0
Preferred Stock (%/yr)	na	4.1	na
Debt (%/yr)	4.0	4.8	9.0
Weighted Cost of Capital (%/yr)	4.0	6.4	13.9
Tax Assumptions			
Income Tax Rate (%)	na	38	38
Tax Recovery Period (yr)	na	15	15
Investment Tax Credit (%)	na	0	0
Book Life (yr)	na	30	30
Annual Fixed Charge Rate (%)	5.8	12.3	20.8

[a] Includes interest during construction.

low cost of capital associated with customer financing of plant capital costs through advanced payments for services for both the THORP and UP3 reprocessing facilities. Following are examples of reprocessing prices:

- The OECD/NEA study developed a levelized reprocessing cost of 720 ECU/kgHM for their reference case, which used a 5% discount rate. They assumed a conversion rate of 1 $/ECU, but the committee applied a rate of 1.07 ECU/$, consistent with the committee's use of average rates for the period 1983 to 1992. This results in a unit reprocessing cost of $770/kgHM, similar to the cost of $810/kgHM developed for a government-owned plant.
- A private communication among Cliff Weber, Department of Energy, Headquarters and European representatives (from both the United Kingdom and France) that was based on OECD input reported prices of $1,400/kgHM for the first 10 years of operation and $600 to $700/kgHM beyond 10 years of operation. The weighted average of these prices over 30 years is $870 to $930/kgHM.
- Suzuki (1994) quoted a unit price of ¥200 million per metric ton for reprocessing services from the Rokkashomura reprocessing plant in the period from 2000 to 2020. At the current yen-to-dollar currency conversion rate, this is equivalent to approximately $1,800/kgHM. • COGEMA confirmed a unit price of FF 6,000/kgHM ($1,250/kgHM) for the first 10 years of UP3 operation (*Nuclear Fuel*, 1987). This price includes waste vitrification and interim storage. It was indicated that after the year 2000, once the plant investment has been largely amortized, the cost might be reduced by 30-40% due an expected increase in plant throughput and the reduced charge for depreciation.
- British Nuclear Fuel, Ltd., indicated it offered German utilities unit price of DM 1,250/kg ($670/kgHM) to reprocess their fuel in THORP.

The committee recognizes that reprocessing prices are not the same thing as reprocessing costs, especially in an environment where there are currently only two reprocessing plants offering commercial reprocessing services. Also, the quoted prices reflected the higher cost of reprocessing services during the initial 10-year amortization period. However, comparing calculated costs with reported prices for government ownership provides a measure of whether derived costs are credible as compared with prices charged in the marketplace. Our estimated unit cost of $882/kgHM (levelized over the assumed plant life of 30 years) for a government-owned reprocessing plant falls within the price range of $600 to $1,400/kgHM reported for actual government/corporation-owned plants.

In contrast to reprocessing prices reported for THORP and UP3, unit costs reported in most reprocessing plant studies are typically much lower, ranging from about $237 to $600/kgHM. A study by Gingold et al. (1991), *The Cost of Reprocessing Irradiated Fuel from Light Water Reactors: An Independent Assessment*, reported unit reprocessing prices of $237/kgHM for a government-owned facility and $489/kgHM for a privately owned (nonutility) facility. The GE study for the ALMR program was based on a very large (2,700 MTHM/yr) reprocessing facility. The size of this facility is unprecedented for a commercial reprocessing plant. GE did not report unit reprocessing costs, but using their capital cost of $5,976 million, their constant-dollar annual fixed-charge rate (utility owner) of 9.246%/yr, their annual operating cost of $114 million, and an annual plant throughput of 2,700 MTHM results in a unit reprocessing cost of $247/kgHM. The unit cost for reprocessing only would be even lower after deleting the capital charges and operating-related costs associated with the fuel fabrication facility.

In a 1983 paper, Wolfe and Judson noted that the estimates of LWR unit reprocessing costs in constant dollars increased substantially from 1975 to 1983 (Figure J-2). Figure J-2 also provides the current estimates of the unit costs for reprocessing plants constructed in the United States, which are derived from estimated costs for contemporary plants in the United Kingdom, France, and Japan. Financial parameters were applied for a private venture in the United States, assuming optimistically that the financing is characteristic of a low-risk project in the chemical industry. The unit costs reflect an inflation-free economy. The unit costs estimated for these three sources fall on extensions of the band of reprocessing costs shown by Wolfe and Judson, but at a level several-fold higher. What may be financially valid for a government-owned European plant that is financed with customer prepayments for reprocessing services, and that has relatively low annual charges on capital investment, is not necessarily applicable to the same plant constructed by private industry in the United States.

Also shown in Figure J-2 are two recent estimates by the ALMR project of costs for reprocessing LWR fuel, one for an aqueous plant (Taylor et al., 1991) and the other for a pyrochemical process (Chang, 1993). Each plant has a throughput of 2,700 MTHM/yr, with high-yield recovery of all actinides and volatile fission products. The estimated unit costs for these two processes vary from $500/kgHM to $350/kgHM. The latter is about eightfold less than the estimated cost for the UP3 plant in France and the new Rokkashamura plant in Japan, neither of which is designed for high-yield recoveries.

Also shown in Figure J-2 are the unit costs estimated in 1991 (Gingold et al., 1991) for aqueous reprocessing and pyrochemical processing. The Stoller estimates are near those made by GE, but they are substantially below the costs for the new British, French, and Japanese reprocessing plants.

Based on these comparisons, there is reason to question the validity of all the recent U.S. estimates for the cost of reprocessing LWR spent fuel. The estimates for aqueous plants are far below the costs inferred from the European and Japanese benchmarks. Since it was difficult to come up with reasonably accurate estimates for the one process for which some information is available, it is questionable that reliable estimates could now be made of the pyrochemical process for LWR spent fuel, which is in a relatively primitive stage of development.

Comparison with Oecd/Nea Breakeven Reprocessing Price

The 1993 OECD/NEA study presented levelized LWR fuel-cycle costs for both the reprocessing and direct disposal options for the back-end of the fuel cycle. The levelized cost (mills per kilowatt-hour) of the reprocessing option was found to be approximately 12% higher than the cost of direct disposal.

NOTES: All estimates are for aqueous reprocessing, except Argonne National Laboratory's estimate for high-recovery pyrochemical reprocessing. Estimates by General Electric and Stoller include TRUEX additions for high recovery of actinides. The capital charge rate is for a low-risk private venture in the United States in an inflation-free economy.

Data shown in small circles are from Wolf and Judson (1983). Results of this study are for PUREX aqueous reprocessing based on data from France □, Japan △, and the United Kingdom ○.

U.S. Private Financing

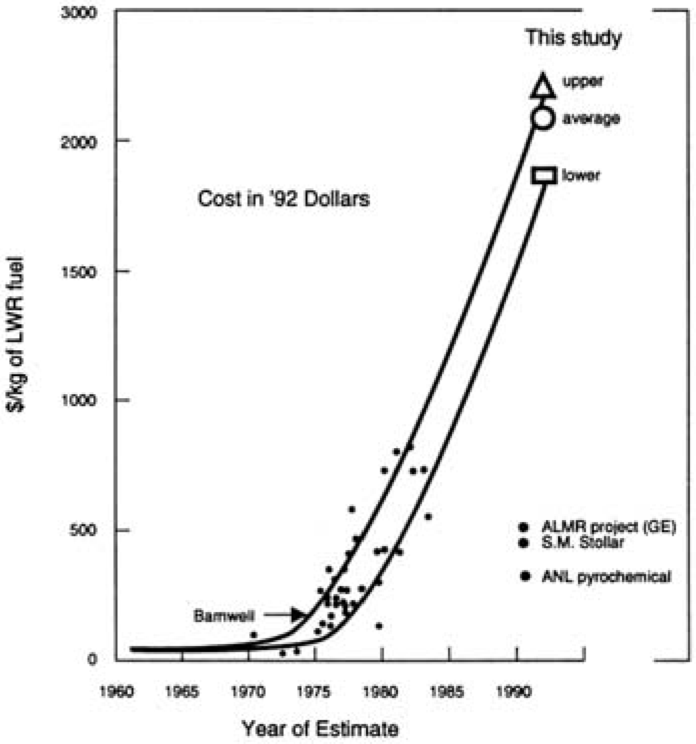

FIGURE J-2 Current estimates of the unit costs for reprocessing plants constructed in the United States.

A reprocessing cost of 720 European currency unit (ECU)/kgHM was used in the analysis, which is equivalent to $720/kgHM, assuming a long-term currency exchange rate of 1 ECU/$ assumed by the OECD/NEA. The breakeven reprocessing unit cost would be $387/kgHM for a 5% discount rate and $327/kgHM for a 10% discount rate. Although it was not the purpose of the Committee on Separations Technology and Transmutation Systems to evaluate the relative economics of the reprocessing and direct disposal options, the very low breakeven cost compared with the costs developed in the study by the committee is indicative of the challenge in judging whether TRU burning is an economical alternative to high-level waste disposal.

Reprocessing-Cost Sensitivity Studies

Sensitivity studies of unit reprocessing costs were performed around the reference cases presented above to illustrate the impact of changes in assumed plant capital and operating cost, financing rate, financial structure, and currency exchange rate. A plant throughput of 900 MTHM was used in all cases.

Figure J-3, J-4 and J-5 illustrate the sensitivity of unit reprocessing cost to variations in capital and operating cost for government, utility, and nonutility private-venture financing assumptions. Figure J-6 illustrates the effect of varying the debt-to-equity ratio for the privately financed case. The unit reprocessing cost decreases rapidly as the debt-to-equity ratio is increased. This simplified analysis ignores the likely increase in the interest on debt capital that would result from the increased project financial risk associated with the higher debt-to-equity ratios. Figure J-7 shows the sensitivity of unit reprocessing cost to return on equity capital and the associated impact of corporate income taxes, and Figure J-8 illustrates the sensitivity of unit reprocessing cost for a government-owned plant to changes in the currency exchange rate between British pounds and U.S. dollars.

Potential Savings For Nth-of-a-Kind Plants

Because centuries of ALMR operation would be required to achieve the reduction in actinide inventory proposed by the ALMR project, additional reprocessing plants would be required as the earlier plants reach the end of their design life, even without further growth in the total amount of installed nuclear power generating capacity. The capital cost of follow-on (replicated) plants would be less as a result of savings in design costs and the learning experience from designing and constructing the first unit. While the committee did not attempt to quantify potential Nth-of-a-kind cost savings, the 25 million man-hour engineering effort quoted by COGEMA for UP3 provides a basis for quantifying design cost savings.

Assuming U.S. engineering costs of $65/hr, the cost of designing the first plant would be $1,620 million. If a standardized, prelicensed facility could be replicated for only one-third to one-half of this amount, total capital cost could be reduced by approximately $500 to $1,000 million.

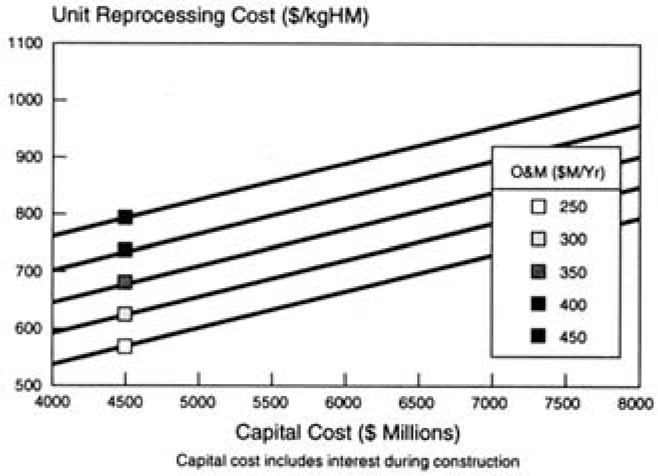

FIGURE J-3 Capital/Operating-Cost Sensitivity for a Government-Owned Reprocessing Plant.

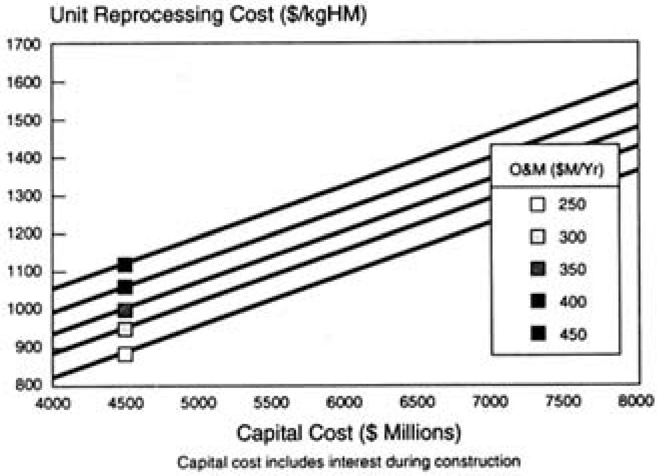

FIGURE J-4 Capital/Operating-Cost Sensitivity for a Utility-Owned Reprocessing Plant.

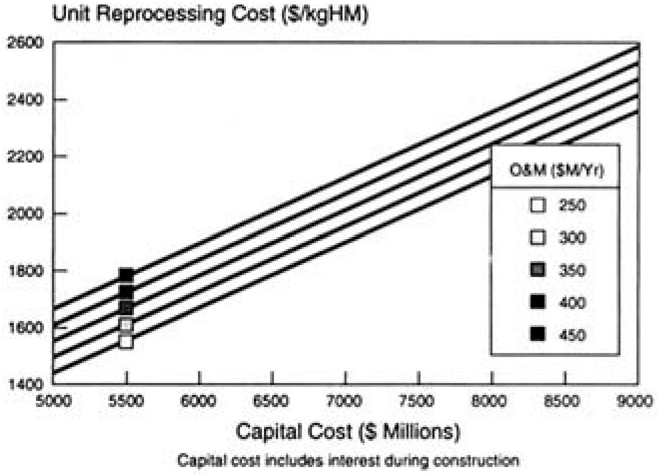

FIGURE J-5 Capital/Operating-Cost Sensitivity for a Privately Owned Reprocessing Plant.

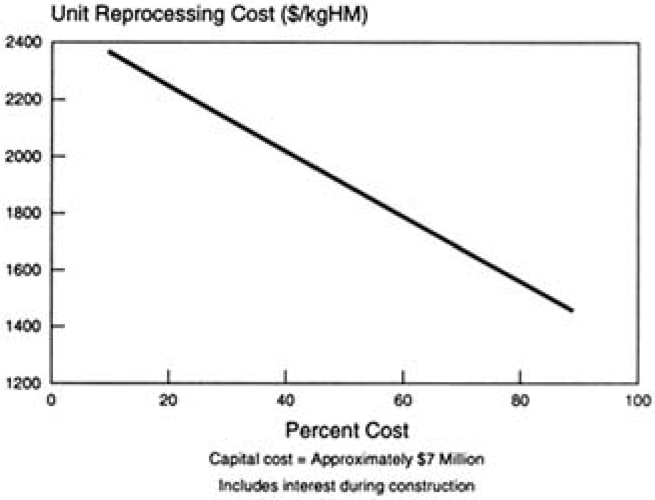

FIGURE J-6 Debt/Equity-Ratio Sensitivity for a Privately Owned Reprocessing Plant.

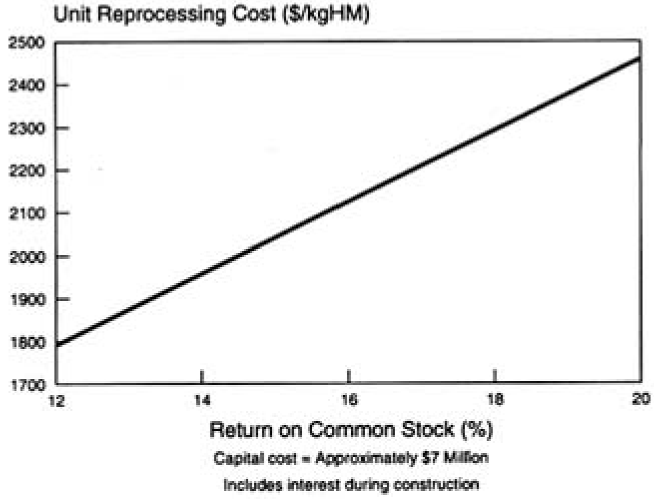

FIGURE J-7 Sensitivity to Cost of Equity Capital for a Privately Owned Reprocessing Plant.

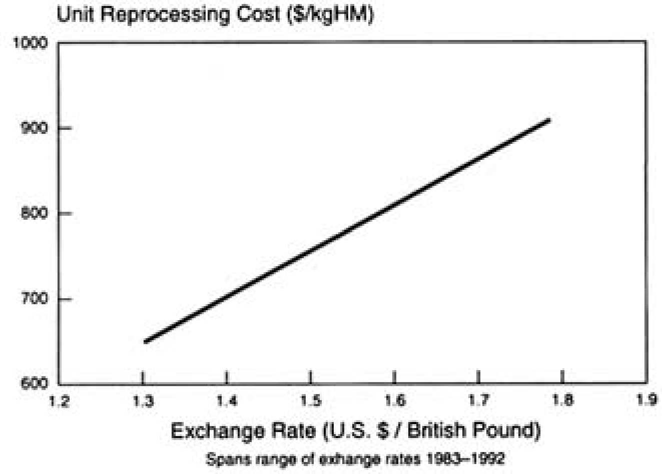

FIGURE J-8 Sensitivity to Currency Exchange Rate of a Government-Owned Reprocessing Plant.

Threshold Cost of LWR Fuel Reprocessing For ALMR Use

The ALMR project proposes to start and refuel future ALMRs using plutonium and other TRUs recovered from LWR spent fuel so as to reduce the quantity of transuranics that would otherwise go to a geologic repository. The project also proposes to use TRUs from LWR fuel for reloads of the nonbreeding ALMRs. However, if the cost of reprocessing LWR spent fuel is too great, it will be less expensive to start first-generation ALMRs with virgin uranium enriched to approximately 30-40% ^{235}U.

If start-up with enriched uranium does not prove to be too expensive for an ALMR to be economically competitive with alternative energy sources, this could be a viable approach to introducing ALMRs for generating electric power. However, such ALMRs would not be a viable means for transmutation of TRUs in LWR spent fuel. Assuming a uranium ore cost of $30/lb of U_3O_8 and a separative work cost of $80/kg separative work unit (SWU) available on the international market, the threshold cost of LWR reprocessing was estimated by Argonne National Laboratory (ANL) (Chang, 1992) to be $350/kgHM. This should also include any cost increment for fabricating ALMR fuel from TRUs, over and above the cost of fabricating makeup fuel from enriched uranium. If the reprocessing/refabrication cost exceeds $350/kgHM, it will be cheaper (and easier) to start and refuel with enriched uranium. There would then be no economic fuel-cycle incentive for the transmutation ALMRs to be started and fueled with actinides from LWR spent fuel to aid the disposal of waste from LWRs.[3]

On this basis, ANL established a target for its new development of pyrochemical processing of LWR spent fuel to cost less than $350/kgHM. ANL now estimates that the cost of pyrochemical reprocessing of LWR spent fuel will be $350/kgHM. The project assumes that the reprocessing facility will be centralized, high throughput, and financed by a private venture typical of low-risk ventures in the chemical industry. Comparing the threshold value of $350/kgHM with the estimated unit cost of a large private-venture reprocessing plant presented above in *Estimated Unit Costs of LWR Reprocessing*, the ANL IFR concept must be able to reduce the unit cost by a factor of about six below that based on contemporary costs for large-scale aqueous reprocessing. The disparity in estimated unit costs would be even greater if the reference aqueous facility were modified by adding TRUEX to obtain high recoveries for transmutation. This is a forbidding target, even if the expected large diseconomy of the small scale of the integrated pyrochemical reprocessing is ignored. An independent estimate made in 1991 by S.N. Stoller (see Gingold et al., 1991) for EPRI concluded that unit reprocessing cost for an investor-owned plant for pyrochemical reprocessing of LWR spent fuel would be 57% greater than that for an aqueous reprocessing facility of the same throughput.

DOE expects the ALMR program to establish technical feasibility by 1995. Technical feasibility must include some measure of practicability. How the development program is expected to contribute to some resolution of these large uncertainties and disparities in cost

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[3] These estimates refer only to the costs of alternative means of starting and refueling an ALMR. Studies by Oak Ridge National Laboratory show that even for a cost of $350/kgHM for reprocessing LWR fuel it would be more economical to build and operate a once-through LWR instead of ALMRs, absent a federal subsidy to encourage waste disposal.

estimates for pyrochemical reprocessing by 1995, or even later, in a facility operating at far lower throughput than that now projected for the LWR spent-fuel reprocessing, and not under NRC regulations, has not been explained.

ANL Estimates of the Cost of Pyrochemical Reprocessing of ALMR Recycle Fuel

The ANL IFR concept that a reprocessing plant for ALMR recycle fuel could be economically colocated with an ALMR power plant is challenged by cost analyses reported in GE's 1991 fuel-cycle assessment for the ALMR project (Taylor et al., 1991). The estimated fuel-cycle cost is due to the costs of reprocessing and fabrication, less a credit of 2.0 mills/kW·h for uranium recovered in reprocessing LWR spent fuel, the uranium to be sold for reuse in fabricating LWR fuel. The calculated fuel-cycle cost for a single colocated IFR, with a nominal throughput of 20 MTHM/yr, is 8.9 mills/kWh in 1991 dollars. This is compared with an estimated fuel cost of 5.4 mills/kWh if a centralized reprocessing facility is used to reprocess ALMR fuel discharged from eight 3-block ALMRs, at a throughput of 200 MTHM/yr. A considerable economy of scale would evidently result from a centralized facility.

GE reported the following costs for the fuel-cycle facilities:

Location	Colocated	Central
Nominal throughput (MTHM/yr)	20	200
Facility equipment cost (first-of-a-kind), $ millions	105	518
Annual manpower (*n*th-of-a-kind), $ millions/yr	10.4	32.8
Operating consumables (*n*th-of-a-kind), $ millions/yr	17.4	143

The estimated capital cost of the recycle facility is estimated to increase by a factor of nearly five for a tenfold increase in plant throughput, which is equivalent to the 0.69 power of the plant throughput. The data in the ALMR project's 1991 fuel-cycle report show considerable economic penalty associated with the small scale of colocated IFR facilities.

The ALMR project's 1991 report also shows an economic penalty of about 0.3 mills/kW·h for recycling all TRUs for transmutation to achieve low inventories for waste disposal. ANL's pyroprocessing/fabrication system is to be remotely operated and maintained. One of the best U.S. benchmarks for sizing equipment for remote operation and maintenance of high-activity reprocessing systems is the 1981 detailed independent study by Bechtel (Jones, 1981) of a proposed hot experimental facility for processing fast-breeder fuel, sponsored by DOE through the Oak Ridge National Laboratory. Although the process is aqueous instead of pyrochemical and the throughput is higher, it appears that the volume of process cells is dominated by the size of the remote fabrication equipment. The process equipment for the hot experimental facility is divided into two main process cells, arranged in parallel and interconnected with a decontamination and maintenance cell. The two process cells are about 40 feet wide by 610 feet long by 80 feet high, for a total volume of about 2 million cubic feet. A review of the layout shows that the volume is dominated by the equipment for remote manipulation and maintenance.

Based on the Bechtel study, it appears that ANL may need a more thorough engineering analysis of the complexity and size of remote manipulation and maintenance operations.

Historical Cost Growth on Projects Employing Advanced Technologies

The ALMR project has adopted pyrochemical separations for reprocessing ALMR discharge fuel and for reprocessing LWR spent fuel. New and improved technologies are involved. The technical and economic challenges are enormous, particularly for the pyrochemical reprocessing of LWR spent fuel. Reprocessing LWR fuel is the critical early-lead item for deployment of the ALMR systems according to DOE's proposed schedule.

The RAND Corporation conducted several studies designed to identify the principal factors that caused the substantial capital cost overruns and performance shortfalls characteristically experienced on pioneer projects. These studies were intended to enable project planners to make more realistic estimates of likely cost growth in moving a new technology from the research and development stage through the design, construction, and operation of a production-scale facility.

The following are some of the major conclusions of the RAND report (Merrow et al., 1981):

- Severe underestimation of capital costs is the norm for *all* advanced technologies.
- Capital costs are repeatedly underestimated for advanced chemical process facilities, just as they are for advanced energy process plants. Furthermore, the performance of advanced energy plants constantly falls short of what was predicted by designers and assumed in financial analyses.
- Greater than expected capital costs and performance shortfalls not anticipated by conventional estimating techniques can be explained in terms of the characteristics of the particular technology and the amount of information incorporated into estimates at various points in project develop.
- Most of the variation found in cost-estimation error can be explained by (1) the extent to which the technology departs from that of previous plants, (2) the degree of definition of the project's site and related characteristics, and (3) the complexity of the plant. • Most of the variation in plant performance is explained by the measures of new technology and whether a plant produces solid materials.

Nuclear fuel reprocessing plants using aqueous technology have experienced such cost growth and performance shortfalls as well. Despite the extensive experience base acquired during the processing of spent fuel from defense production reactors, application of PUREX technology to commercial reactor-fuel reprocessing proved to be technically challenging. Current unit prices for LWR reprocessing services are nearly an order of magnitude greater than the optimistic estimates of the early 1970s. Now that the technology has matured, however, costs should be relatively stable. Part of the increase in cost is attributable to much tougher regulations that were not in place when some of the early LWR plants (e.g., Barnwell and Exxon in the United States) were designed. ANL had to upgrade the EBR-II LWR reprocessing facility prior to performing its demonstration in the early 1990s.

The limited experience with pyrochemical reprocessing of LWR fuel as proposed by the ALMR program offers no compelling argument that current cost estimates will not increase as detailed designs emerge or that such reprocessing would be less costly than conventional aqueous reprocessing of LWR fuel, or even less costly than a PUREX facility augmented with the aqueous TRUEX process to achieve high-yield recovery of TRUs for ALMR transmutation.

Pyrochemical reprocessing represents a substantial departure from aqueous (PUREX) technology. It is characterized by first-of-a-kind components, remote maintenance, solids handling, etc. Many of the factors that were identified in the RAND study as having caused cost overruns and performance shortfalls on previous projects would appear reasonable.

Summary and Conclusions

The transmutation of actinides and fission products recovered from spent fuel offers the potential for improving the technology for long-term disposal of radioactive waste. Successful implementation of this integrated reprocessing/irradiation strategy would require a major financial commitment to the development, design, construction, and operation of a series of reprocessing plants and transmutation reactors both of which are based largely on as-yet unproven technology.

The most reliable cost data for reprocessing services is that associated with aqueous processing of spent LWR fuel. Pyroprocessing technology, which is being considered by both ANL and GE for reprocessing ALMR fuel, has not yet evolved to the point where estimates of capital and operating costs can be prepared with sufficient confidence to estimate future unit prices of reprocessing services. Moreover, it is by no means certain that pyroprocessing will prove more economical than conventional aqueous reprocessing for which the technology is now relatively mature.

The LWR reprocessing capital costs used in this study to develop life-cycle unit reprocessing costs were derived from information reported for the THORP, UP3, and Rokkashomura plants, all of which have an annual throughput of 800 to 900 MTHM/year. Reported capital costs ranged from $5,300 to $7,400 million for these plants. When scaled to a common annual throughput of 900 MTHM/yr, these costs are typically two to three times greater than the capital costs presented in recent reprocessing plant studies for new U.S. plants.

Very little information is available in the open literature with regard to operating costs for the above three plants. The committee adopted the operating cost presented in the 1993 OECD/NEA study of the nuclear fuel cycle, which is $386 million/yr when converted to 1992 U.S. dollars.

Unit reprocessing costs were calculated for government-owned, utility-owned, and privately owned reprocessing plants. The ground rules assumed for each case are those shown previously in Table J-5. The capital and operating costs, developed from OECD/NEA, were

$5,770 million ($6,160 to $7,320 million with interest during construction) and $374 million/yr, respectively. Plant throughput was 900 MTHM/yr.

The resulting levelized unit reprocessing costs are

Owner/Operator	Unit Cost ($/kgHM)
Government	810
Utility	1,330
Private venture	2,110

Based on the above comparisons, there is good reason to question the validity of all the recent U.S. estimates for the cost of reprocessing LWR spent fuel. Given that the estimates for aqueous plants ranging from $237 to $600/kgHM are so far below the costs inferred from the European and Japanese benchmarks, it is questionable that more reliable estimates could now be made of the pyrochemical process for LWR spent fuel, which is in a relatively primitive stage of development.

The committee concludes that if it were cost effective, government ownership of reprocessing and transmutation facilities would be the only realistic alternative. The government is ultimately responsible for long-term disposal of HLW and is the only institution potentially capable of overcoming the complex political/institutional issues associated with reprocessing and transmutation. This responsibility may be difficult to achieve in the United States considering that, historically, government opposition to reprocessing has been the principal deterrent to its implementation outside the defense materials production facilities. Government ownership is nevertheless the logical choice when considering the relative cost under government, utility, and private (nonutility) financing arrangements. However, studies by DOE's ALMR project emphasize private ownership and financing of the ALMR reactors and associated reprocessing/fabrication facilities, which the committee considers an unrealistic expectation.

Achieving the high degree of confidence in the cost and performance estimates that is necessary for a private corporation (including a utility) to commit the capital resources to proceed with an transmutation demonstration project would require a very extensive development and demonstration program, likely funded primarily by the government. A government project might be able to accept a higher level of risk, but the enormous magnitude of the commitment over 100 years or more would challenge even the federal government's ability to undertake such a project.

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Appendix K

Summary of International Separations and Transmutation Activities

During the last few years, there has been a substantial increase in the number of countries and international organizations undertaking programs related to investigation of one or more aspects of separations and transmutation (S&T). The purpose of this appendix is to provide a summary of the most significant activities. Except where specifically noted, the information presented in this section is based on a report prepared by the Electric Power Research Institute and its contractors (EPRI, 1991) and papers presented at the National Research Council Symposium of Separations Technology and Transmutation Systems (National Research Council, 1992). For recent information on technological details of international S&T activities, the reader is referred to (OECD/NEA 1990, 1992).

The section in which a particular activity is discussed is somewhat arbitrary at times. The complex web of collaborative international activities and organizations makes definitive assignment of activities to specific installations or nations very difficult or impossible.

Programs Of Individual Nations

China (OECD/NEA, 1990)

China, though not in possession of any operating nuclear power reactors, has been reprocessing spent fuels from its other reactors since the 1960s. During this time, three processes have been developed to separate actinides from high-level waste (HLW): reverse TALSPEAK, a process in which the $Am^{(III)}$ + $Ln^{(III)}$ elements are extracted in an organic phase by di (2-ethylhexyl) phosphoric acid (HDEHP). Research activities to optimize the reverse TALSPEAK and DHDECMP process are ongoing, along with work on separation of platinum-group metals and cleanup of degraded solvents. Hot demonstration of actinide extraction is planned. Transmutation is not yet being studied.

Czech and Slovak Federal Republic (OECD/NEA, 1990)

The Institute of Nuclear Research in Rez, Czech and Slovak Federal Republic (CSFR) has synthesized a new extractant, cobalt dicarbollide, under the sponsorship of the Radium Institute in Leningrad. This extractant shows promise for separation of radiocesium and, when mixed with polyethylene glycol, for separating radiostrontium from highly acid solutions. Laboratory work has been conducted in the CSFR, and hot testing at the Radium Institute. The work is still in the research stage because of difficulties with low product concentrations, the use of nitrobenzene as a diluent, and the high cost of the extractant. Work is currently suspended because of the recent political change in the former Soviet Union.

France

France is committed to nuclear fuel reprocessing and actinide recycle as a matter of national policy, and these activities are presently practiced domestically and as part of international commerce. During the last few years, France has initiated work to establish a geologic repository for the HLW and other long-lived wastes resulting from their nuclear activities. Recognizing the controversial nature of such facilities, the French legislature adopted a long-term approach to repository development. In addition, specific legislation was passed in 1991 that, among other things, called for continuing study of S&T with a final assessment to be provided within 15 years.

The French have been relatively explicit about their rationale for pursuing S&T (National Research Council, 1991). They currently believe that the reduction in long-term repository risk is not large enough to justify S&T, primarily because the absolute risk values are below levels deemed acceptable. However, they acknowledge sufficient uncertainties in the risk calculations (e.g., concerning actinide solubility limits, colloids, and complexes) that continued examination of S&T is warranted.

The French program to investigate S&T is called SPIN. It comprises two major subparts: PURETEX and ACTINEX. The former is a shorter-term program to increase the recovery of plutonium, separate and recycle neptunium, and reduce the amounts of intermediate-level waste (roughly equivalent to transuranic [TRU] waste in the United States), primarily within existing production facilities. The latter is a longer-term effort involving enhanced separation of all TRU elements, separation of long-lived fission products, and recycle of the TRUs and fission products to transmutation devices. This effort would involve new facilities.

Separation studies are heavily focused on aqueous systems, although nonaqueous processes are being considered. The behavior and control of neptunium in PUREX processing is being closely studied for near-term applications. Of particular interest, the French are examining the use of diamides as alternatives to the reagents employed in the U.S.-developed TRUEX process (i.e., carbomoyl methyl phosphine oxide [CMPO]). Nitrogen and sulfur donors are being studied for the purpose of separating $Am^{(III)}$ from lanthanides, a traditionally difficult separation. Electro-oxidation of actinides to accomplish this separation is also being considered.

Transmutation efforts are focused on thermal and fast nuclear reactors, although some consideration is to be given to accelerators. Some fundamental physics data is being obtained (e.g., actinide cross sections), and test pins are being irradiated in various French reactors.

The budget for SPIN is approximately $600 million over 15 years, which includes funding for new research and development facilities at Marcoule. The 1992 budget was about $17 million. In both cases, there is somewhat of an emphasis on PURETEX. The effort for 1991 was 20 person-years. This may expand to 40 to 50 person-years within a 5-year plan.

Germany

Germany is not now reprocessing fuel and has no immediate prospects for doing so. However, study of reprocessing and S&T continues for the future. Of particular interest is study of a single-cycle aqueous process for reprocessing spent fuels. These activities are centered at Karlsruhe, at the Institute of Nuclear Research.

India (OECD/NEA, 1990)

India is conducting studies of the nuclear physics aspects of the minor actinides that are directed at processing and compiling a database that would support accurate reactor-physics calculations.

Italy (OECD/NEA, 1990)

A number of Italian radiochemical facilities are in the process of being decommissioned due to budget cuts. The Italians, in a program partially funded by the Commission of the European Communities (CEC), are pursuing separations technology to enable them to better manage their wastes. Multiple partitioning processes for actinides, technetium, strontium, and cesium from various waste streams have been identified and tested on simulated wastes for the purpose of identifying the preferred processes. The next phase calls for hot continuous testing of the preferred process in a small pilot plant at the Joint Research Center at Ispra, which is scheduled to be completed in 1995.

Transmutation studies have been historically performed on thermal and fast neutron reactors. More recently, the Italians have performed confirmatory reactor physics calculations on the aqueous accelerator transmutation flow of wastes (ATW) and compared them with Los Alamos National Laboratory (LANL) calculations (OECD/NEA, 1992), with some significant differences being noted.

Japan

The Japanese are committed to reprocessing as a matter of national policy and have the most comprehensive and organized S&T program of any nation or organization. Japan's Option Making Extra Gains from Actinides and Fission Products (OMEGA) program—started in 1985 and formalized in 1987—is a comprehensive, long-term effort involving data acquisition and evaluation, study of partitioning schemes, and transmutation of nuclides in reactors or by using accelerators. OMEGA funding is nominally about $10 million per year, although it is not clear how much "work in kind" is being provided by the participating organizations. The OMEGA program is only summarized briefly here. A paper given at the Separations and Transmutation

Systems (STATS) Symposium (National Research Council, 1992) provides an excellent, detailed overview of the OMEGA program.

The Japanese program involves multiple organizations, including the Japan Atomic Energy Research Institute, the Power Reactor and Nuclear Fuel Development Corporation, and the Central Research Institute of Electric Power Industry.

Transmutation studies are directed at the use as a transmutation device of metal-and oxide-fueled fast reactors (liquid-metal reactors [LMRs]), proton accelerators (i.e., spallation neutrons), or electron accelerators (i.e., photons). The predominant emphasis is on oxide systems as fuel forms, but metal fuels and pyrochemical processing are also being considered.

Partitioning studies involve significant efforts on both aqueous and nonaqueous technologies, although the former are dominant. Partitioning studies are focused on recovery and recycle of long-lived toxic radionuclides as well as recovery and reuse of useful (e.g., noble) metals.

The Japanese appear to be approaching this issue from a collaborative standpoint, having established working agreements with multiple countries. Within the United States, they are known to have working arrangements with Oak Ridge National Laboratory, Battelle, Argonne National Laboratory (ANL), and Rockwell International.

Korea (OECD/NEA, 1990)

The Republic of Korea has embarked on a three-phase, 10-year program to investigate S&T technologies and selected isotope separations related to waste management. A variety of separations technologies are being examined for separating actinides and selected fission products from wastes. The initial step involves literature surveys and laboratory studies to elucidate the nature of the radioelements of interest, which are scheduled to be completed by 1993. This will be followed by cold laboratory studies through 1996 and the specification, design, and operation of a pilot-scale process by 2000.

The separation of long-lived ^{107}Pd from radiopalladium using laser isotope separation techniques is being studied. This work is currently in the basic research stage.

Transmutation of actinides using proton accelerators has temporarily been selected as the preferred method, and this technology is being studied. Tests are expected to be conducted collaboratively with other nations.

Netherlands

The Dutch government has requested that the Netherlands Energy Research Foundation study actinide transmutation. The foundation is currently studying the use of thermal reactors, fast reactors, and accelerators in this application, especially with regard to the nuclear physics of the transmutation process.

Russia

Russia has a long-standing and continuing interest in multiple aspects of S&T going back decades to when cesium and strontium were separated from defense HLW. Research and development activities continue concerning transmutation studies, nuclear physics (cross section libraries), partitioning technology (aqueous and nonaqueous), actinide-recycle fuel fabrication, special actinide burning cores for fast breeder reactors, use of military/defense reactor systems (e.g., lead/bismuth-cooled reactors) for transmutation, and special burner reactors. Extensive work has been undertaken on accelerators for a variety of applications, and there is apparent interest in their application to transmutation systems.

Sweden

A modest effort is underway in the Chalmers University of Technology (CTH), Götenborg, to examine aspects of S&T. Some transmutation calculations have been performed. Experimental and calculational study of actinide partitioning has been performed on cold and hot solutions based on the use of tributyl phosphate and di (2-ethyl hexyl) phosphoric acid extractants in what is designated the "CTH Process."

Switzerland

The Paul Scherrer Institute has conducted calculational studies related to actinide transmutation via spallation. Irradiation of small samples is proposed to validate the calculations.

United Kingdom

The United Kingdom view of S&T is that it may be costly and has not been shown to be desirable. The United Kingdom is continuing to monitor and evaluate worldwide developments related to S&T with the expectation that such processes will prove to be undesirable after further evaluation.

As is France, the United Kingdom is dedicated to fuel reprocessing as a matter of national policy and international commerce. The United Kingdom approach is to perform research and development to improve aqueous reprocessing which in turn will improve its cost-effectiveness of the reprocessing. The cost-effectiveness is to be balanced against increased risk by using a philosophy of "best available technology not entailing excessive cost."

Research and development efforts by the British that may relate to S&T include the development of technologies to sharply reduce the cost of reprocessing. These include adoption of centrifugal contractors; study of technetium behavior during aqueous processing to prevent it from partially following product streams; and consideration of "radical PUREX," in which the entire fuel pin is dissolved (to reduce head-end costs), a simplified (perhaps one-cycle) extraction flowsheet is used, and alternative product conversion technologies (e.g., thermal denitration) are employed.

United States

The United States is presently committed to a once-through fuel cycle. However, a number of organizations are undertaking studies related to S&T. These studies are largely the result of initiatives by the individual organizations. An integrated governmental program regarding S&T is absent.

An advanced liquid-metal reactor program (ANL and General Electric) is developing sodium-cooled fast reactors, primarily for use in electricity production. These devices have also been proposed as actinide transmutation devices. Essentially the entire focus is on metal fuels and pyrochemical reprocessing of these fuels. However, oxide fuels and aqueous reprocessing is maintained as a back-up and for potential use in international applications.

LANL has proposed a fluid-fuel, accelerator-based system for use in a number of potentially synergistic applications. These include TRU transmutation, long-lived fission-product transmutation, destruction of plutonium from dismantled nuclear weapons, and production of tritium. The systems involve subcritical transmutation via proton-generated spallation neutrons. Aqueous and molten-salt systems are being examined.

Westinghouse Hanford Co. has put forth the Clean Use of Reactor Energy (CURE) concept regarding S&T. This is more a systems concept for S&T than a specific transmutation device or partitioning technology. It advocates the complementary use of various technologies to achieve S&T, and is primarily based on aqueous/oxide systems.

Brookhaven National Laboratory is pursuing two technologies related to S&T. The first involves the use of accelerators to transmute actinides. In this case, the protons impinge directly on the targets (in the form of standard fuel assemblies), and neutron spallation is secondary. The second technology involves adaptation of a high-flux thermal reactor designed for space applications. This device is based on high-temperature gas-cooled reactor (HTGR) fuel technology and has a high specific power (i.e., transmutation rate) during the in-core period.

International Organizations

The Commission of the European Communities (CEC)

The CEC operates four joint research centers with funding from its member states: Geel (Belgium), Ispra (Italy), Karlsruhe (Germany)—also known as the Institute for Transuranium Elements—and Petten (Netherlands). The Karlsruhe and Ispra sites are the most heavily involved in S&T activities. The CEC is studying various aspects of S&T at the request of the Parliament of the European Community. The work is supported in part by the member states of the

European Community and in part under cost-sharing arrangements with other organizations or states.

Activities at Karlsruhe include:

- basic studies of actinide properties (e.g., single crystals) and californium neutron source manufacture;
- manufacture and postirradiation examination of targets inserted into existing LMRs (e.g., Phénix, Dounrey);
- minor-actinide-fueled LMR core design and safety analysis; and
- study of zirconium-based metal alloy fuels, especially related to the maximum minor actinide content thereof.

Study of S&T has been conducted in Italy under the joint sponsorship of the CEC and the Italian government, and more activities are planned for the future. These are described under the section on Italy above.

International Atomic Energy Agency (IAEA) of the United Nations

The IAEA is primarily a coordination agency, which focuses on the nations that are not part of the Organization of Economic Cooperation and Development (OECD) (i.e., the less industrialized ones) and represents their views in various venues.

A study conducted in the 1970s by the IAEA concluded that there is little incentive to reduce long-term hazards of actinides and fission products by S&T. The incremental cost of introducing S&T was considered unduly high in relation to the prospective benefits. After a period of dormancy—there is now a renewed interest in the subject in some countries—an advisory group meeting was held in 1991 to discuss S&T. The group recommended setting up a scientific advisory body to coordinate the activities of those countries that are a part of the Nuclear Energy Agency of the OECD (OECD/NEA) and those that are not OECD/NEA members. A 5-year program consisting of advisory and technical committee meetings and coordinated research programs has been formulated. IAEA considers S&T as a futuristic option and that S&T cannot avoid the need for long-term, deep geological disposal (IAEA, 1991).

OECD/NEA

The NEA is the arm of the OECD that addresses nuclear issues. The OECD has 23 nations as members, which is larger than the CEC but includes its 12 member nations. The standing committees of the NEA (i.e., development, nuclear science, radioactive waste management) all conduct technical studies using contributions from the member nations (general and proposal-specific) in conjunction with the contribution of work-in-kind from interested nations. The NEA has recognized that S&T is not an alternative to a repository but is a technology worthy of further study for potential longer-term application.

The NEA is actively supporting a program of informal S&T information-exchange meetings initiated by Japan, and additional financial support has been provide by Japan. Fourteen countries and the CEC have identified liaison officers for the information exchange. At the first meeting held at Mito, Japan (OECD/NEA, 1990), the participants concluded that there were benefits to wider international cooperation regarding S&T. Meetings of specialists on specific topics within the S&T rubric have been organized at Karlsruhe, Germany (on fuels and targets), Mito, Japan (on partitioning technology), and the Paul Scherrer Institute in Switzerland (on accelerator-based transmutation).

In addition, the OECD supports other studies that are relevant to S&T. These generally relate to the economic and risk aspects of nuclear fuel cycles and competing energy-production technologies. These aspects are important components of the systems studies necessary to evaluate the S&T concept and alternatives within it.

Observations

- S&T is being studied to varying degrees by most major nuclear nations and international organizations. Japan, France, and the United States are putting considerable efforts into S&T. A number of other nations have more modest efforts. The notable exception is the United Kingdom, which has adopted a "wait and see" posture.
- In all cases, it is clear that S&T is not viewed as a replacement for a repository. However, there is some observed correlation between the current difficulty in establishing a repository (technical or institutional) and the degree of interest in S&T. For Japan, the interest may also be due to a lack of uranium and other energy resources.
- In France and the United Kingdom, the distinction between separations and "process improvement" is becoming blurred. Activities designated as separations are resulting in process improvements, and many aspects of process improvement (or evolution) would facilitate separations.
- No nation has committed itself to deploy S&T technology, although the French are installing provisions to recycle neptunium and have a program with a near-term focus to improve operations in their existing reprocessing plants. A few nations are committed to long-term programs to further develop the technology and make decisions in 10 years or so. The programs of most nations are not committed to a specific agenda, with the timing and nature of decisions to be determined.
- The emphasis on the relatively more difficult separations aspects (as compared with transmutation) has increased from previous episodes of significant work on S&T. Relatively little work is evident on the fabrication aspects of fuels with increased concentrations of minor actinides or long-lived fission products.
- Most programs are in the relatively inexpensive research and development stage. The higher costs attendant to radioactive work at larger scales has engendered a general desire for international collaboration that is much broader than in previous episodes of studying S&T.

- With the exception of that of the United States, most separations research and development is focused on aqueous processes. Transmutation is mostly focused on a variety of fission reactors, although there are significant efforts on accelerator-based transmutation.

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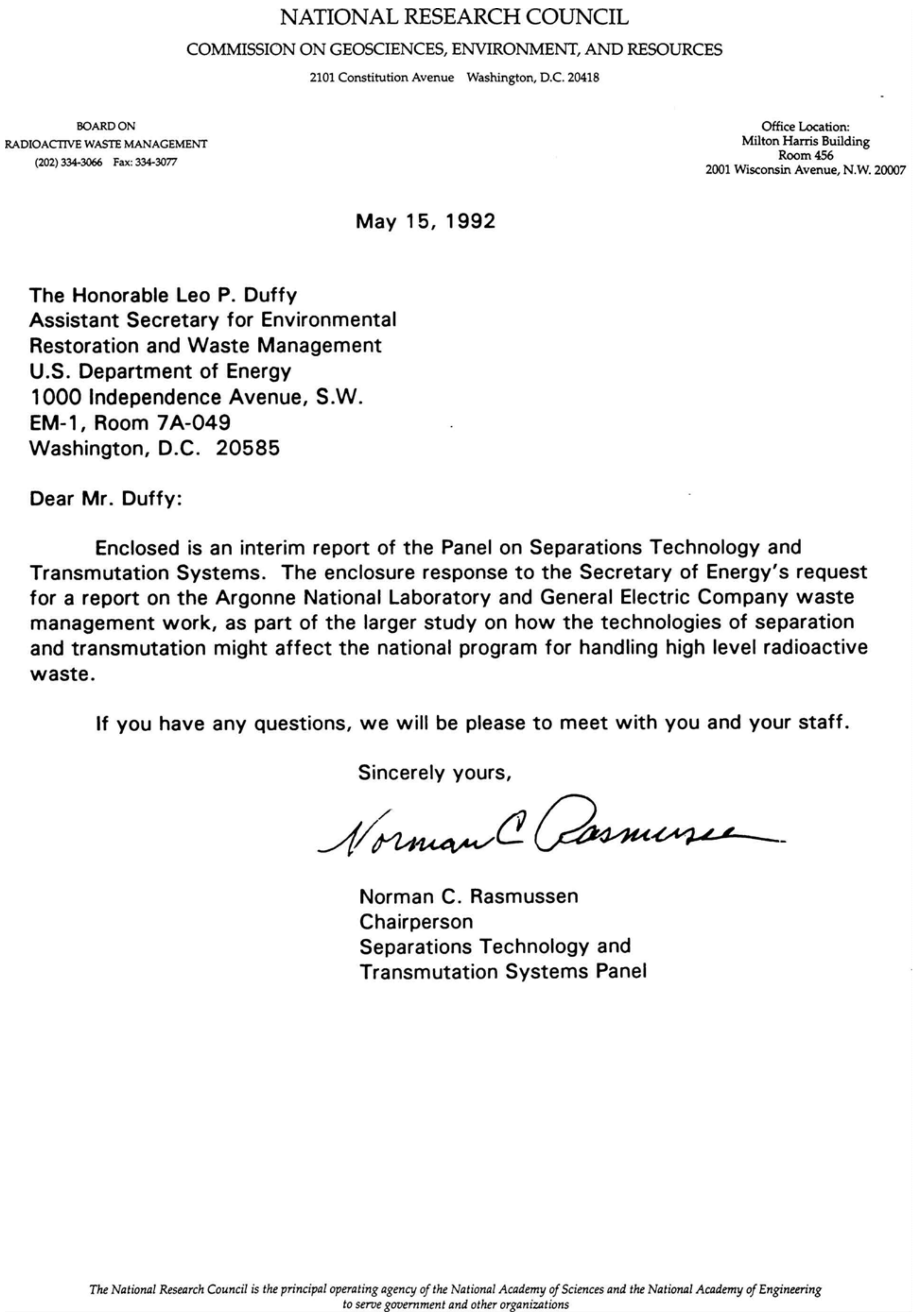
Appendix L

Interim Report of the Committee on Separations Technology and Transmutation Systems

May 15, 1992



NATIONAL RESEARCH COUNCIL
COMMISSION ON GEOSCIENCES, ENVIRONMENT, AND RESOURCES
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BOARD ON
RADIOACTIVE WASTE MANAGEMENT
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May 15, 1992

The Honorable Leo P. Duffy
Assistant Secretary for Environmental
Restoration and Waste Management
U.S. Department of Energy
1000 Independence Avenue, S.W.
EM-1, Room 7A-049
Washington, D.C. 20585

Dear Mr. Duffy:

Enclosed is an interim report of the Panel on Separations Technology and Transmutation Systems. The enclosure response to the Secretary of Energy's request for a report on the Argonne National Laboratory and General Electric Company waste management work, as part of the larger study on how the technologies of separation and transmutation might affect the national program for handling high level radioactive waste.

If you have any questions, we will be please to meet with you and your staff.

Sincerely yours,

Norman C Rasmussen

Norman C. Rasmussen
Chairperson
Separations Technology and
Transmutation Systems Panel

The National Research Council is the principal operating agency of the National Academy of Sciences and the National Academy of Engineering to serve government and other organizations

Interim Report of the Panel on Separations Technology and Transmutation Systems

Board on Radioactive Waste Management
Commission on Geosciences, Environment, and Resources
Energy Engineering Board
Commission on Engineering and Technical Systems
Board on Chemical Sciences and Technology
Commission on Physical Sciences, Mathematics and Applications
May 1992
National Research Council
Washington, D.C.

NOTICE: The project that is the subject of this report was approved by the Governing Board of the National Research Council, whose members are drawn from the councils of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine. The members of the panel responsible for the report were chosen for their special competencies and with regard for appropriate balance.

This report has been reviewed by a group other than the authors according to procedures approved by a Report Review Committee consisting of members of the National Academy of Sciences, the National Academy of Engineering, and the Institute of Medicine.

The National Academy of Sciences is a private, nonprofit, self-perpetuating society of distinguished scholars engaged in scientific and engineering research, dedicated to the furtherance of science and technology and to their use for the general welfare. Upon the authority of the charter granted to it by the Congress in 1863, the Academy has a mandate that requires it to advise the federal government on scientific and technical matters. Dr. Frank Press is president of the National Academy of Sciences.

The National Academy of Engineering was established in 1964, under the charter of the national Academy of Sciences, as a parallel organization of outstanding engineers. It is autonomous in its administration and in the selection of its members, sharing with the National Academy of Sciences the responsibility for advising the federal government. The National Academy of Engineering also sponsors engineering programs aimed at meeting national needs, encourages education and research, and recognizes the superior achievements of engineers. Dr. Robert M. White is president of the National Academy of Engineering.

The Institute of Medicine was established in 1970 by the National Academy of Sciences to secure the services of eminent members of appropriate professions in the examination of policy matters pertaining to the health of the public. The Institute acts under the responsibility given to the National Academy of Sciences by its congressional charter to be an adviser to the federal government and, upon its own initiative, to identify issues of medical care, research, and education. Dr. Samuel O. Thier is president of the Institute of Medicine.

The National Research Council was organized by the National Academy of Sciences in 1916 to associate the broad community of science and technology with the Academy's purposes of furthering knowledge and advising the federal government. Functioning in accordance with general policies determined by the Academy, the Council has become the principal operating agency of both the National Academy of Sciences and the National Academy of Engineering in providing services to the government, the public, and the scientific and engineering communities. The Council is administered jointly by both Academies and the Institute of Medicine. Dr. Frank Press and Dr. Robert M. White are chairman and vice chairman, respectively, of the National Research Council.

Support for the project was provided by the United States Department of Energy.

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Contents

Foreword

At the Department of Energy's request, the National Research Council of the National Academy of Sciences and the National Academy of Engineering has undertaken a study of how the technologies of separation and transmutation might affect the national program for handling high level radioactive waste. To conduct this study, the National Research Council formed the Panel on Separations Technology and Transmutation Systems (STATS) in the fall of 1991, with instructions to issue a final report by July of 1994.

The STATS Panel in its first six months of operation has held a two-day international symposium and has had briefings from the Department of Energy, the Nuclear Regulatory Commission, the Environmental Protection Agency, the General Electric Company, the Electric Power Research Institute, Argonne National Laboratory, Los Alamos National Laboratory, Brookhaven National Laboratory, Hanford Engineering Development Laboratory, and the Yucca Mountain Project. In addition, members of the Panel have made visits to Argonne National Laboratory-East, Argonne National Laboratory-West, Los Alamos National Laboratory, Savannah River, Hanford, and THORP (in the U.K.). This fact-finding effort provided the Panel a large amount of information, not all of it consistent, about these technologies. The STATS Panel is now about to embark on the analysis phase in which we shall draw conclusions about the role separation and transmutation might have in the national high-level radioactive waste program.

Some months ago the Department of Energy asked the Panel for a report at about this time, on any insights we might have gained about the General Electric Company-Argonne National Laboratory concept of actinide burning (fissioning) using fast reactors. It would be inappropriate for us to state conclusions about technologies that we have just begun to study. We can, however, review some of the possible results of actinide burning *if the system works as well as the proponents have contended it will*.

This report should not be considered complete in the sense that all important features have been considered. For example, we need to go into greater detail on mass balances; we must address costs and schedules; defense wastes are only touched upon in passing; accelerator transmutation is not addressed - nor is transmutation in reactors other than the ALMR; foreign programs are not addressed herein; and the important topics of impact on proliferation and other institutional issues are not addressed at all. These issues will be considered in the final report.

The Advanced Liquid Metal Reactor (ALMR) as an Actinide Burner

The Advanced Liquid Metal Reactor (ALMR) concept is being proposed by its designers, the General Electric Company and Argonne National Laboratory, as a fast breeder reactor to generate electricity in the future. Here, we are only concerned with its use as an actinide (specifically, transuranic actinide) burner. For this application the core could be redesigned to reduce the breeding ratio to less than 1. Some proponents say that values as low as 0.2 could be attained (Reference 1), but others assert safety considerations would make the practical limit about 0.6. (Reference 2).

With a breeding ratio of less than 1, the reactor would produce less actinides than it burns, and so with enough burner reactors it would be possible to burn up (i.e., fission) the transuranic actinides that are accumulating in spent light water reactor fuel. If enough of the light water reactor fuel actinides were consumed, the geological repository capacity requirements and the long-term health risks of the repository might be reduced (Reference 3).

A reprocessing plant for spent light water reactor fuel would be required to prepare the light water reactor actinides for burning. The uranium would be separated, and then stored or recycled. Most of the actinides and some fission products (lanthanides) would be made into fuel for the ALMR. The remaining fission products and a small amount of actinides would be removed and disposed of as high level waste in a geological repository. In addition to ALMRs and reprocessing plants for light water reactor fuel, ALMR fuel fabrication plants and reprocessing plants for ALMR fuel would be required (Reference 4). The residual fission product and actinide waste would go to a geological repository. Some suggest removal of cesium and strontium from that waste, and temporary surface or subsurface storage (50 to 100 years) to allow the cesium and strontium to decay to reduce the heat load introduced to the geological repository.

Is a breeder program, viable on its own merits, a prerequisite to an actinide burning ALMR program?

All the people we heard from who commented on this point, including ALMR project participants, felt it made no sense to develop and deploy ALMRs solely for actinide burning. The breeder will be introduced when public policy, licensing, and economic considerations, such as the cost and availability of uranium, justify it. The ALMR proponents and some others felt that if the United States had a healthy breeder program, then it might be possible and justified to modify some or all of the breeder reactors to be actinide burners.

What could be achieved by actinide burning and on what time scale?

Actinide burning greatly reduces the actinides sent to the repository by storing them on the surface in ALMRs and slowly burning them. The fraction of the actinide inventory that could be consumed by this process depends upon the process decontamination factor, reactor fuel-cycle parameters, and length of time of ALMR operation. Based on ALMR project data, including actinide process decontamination factors specified as desirable goals by the ALMR project of

1,000 or more (References 1 and 8), the inventory of actinides that would otherwise accumulate in spent fuel from light water reactors could ultimately be reduced by a few hundred-fold in ALMRs operating continuously at constant power. Transuranic actinide inventory reduction is enhanced by a low breeding ratio, b (so that the ALMR generates fewer new actinides), and by a high process decontamination factor, g (because 1/g is the fraction of transuranic actinides that are lost to waste in each reprocessing cycle). Referring to Figure 1 (from Reference 7, based on Reference 8), we see that, independent of the design parameter b, and for g greater than 1,000, the time required to reach an inventory reduction factor of 10, equivalent to burning 90% of the actinides, would be more than 100 years, the time to reach an inventory reduction factor of 100 would be more than 1,000 years, and the time to reach an inventory reduction factor of 1,000 would be more than 10,000 years. This indicates that the actinide inventory reduction factor that can be achieved in a few centuries is nearer 10 than 100 or 1,000.

A typical ALMR with breeding ratio of 0.62 would require about 30 metric tons of actinides for its initial charge and refueling for a 40-year life (Reference 5). The accumulated actinides in U.S. light water reactor spent fuel by the year 2011 are estimated as about 600 metric tons. Thus, even if light water reactors are phased out, about 20 ALMRs would be required to treat the actinides in U.S. light water reactor spent fuel on hand in 2011. The actinides in those 20 ALMRs at end of life would have to be burned in the next generation of about 10 ALMRs, and so on until the remaining actinides are insufficient to fuel one ALMR; those remaining actinides would go to a repository. If the light water reactor economy persists, many more ALMRs will be required for a longer time.

What would be the effect on the radiation-induced health risks?

The effect of actinide burning on radiation-induced health risks depends on just which isotopes contribute most to long term health risks due to their possible leakage from the repository. The long-lived actinides have a greater potential for radiation-induced health impact than do short-lived fission products after about 300 years, but the long-lived isotopes of technetium and iodine are also of concern. The health risks depend not only on the toxicity of the material but also on how rapidly it spreads into the environment. It turns out under all planned repository conditions that the actinides are much less soluble than the long-lived fission products. Thus, for solubility-limited repository scenarios, the fission products dominate the health effects, and burning of the actinides produces little or no reduction in the health risk. However, in invasive scenarios such as those involving human intrusion, the actinides are expected to dominate risk and a benefit is obtained from a reduction in actinide content of the waste.

If we assume that any ALMR introduced into the electric utility grid would replace a light water reactor, use of the ALMR would allow already-mined depleted uranium or uranium from spent fuel to be used instead of natural uranium. Then the associated reduction in radioactivity from radon and other components in uranium ore would be expected to more than compensate for radioactivity released as a result of reprocessing for the ALMRs (Reference 3). On the other hand, the current ALMR flow sheets show technetium-contaminated uranium as a by-product of pyroprocessing of light water reactor spent fuel (References 1 and 5). Although we have not studied

Ψ is the ratio of (inventory of transuranic actinides at time τ in spent fuel from the LWR at constant power, with no ALMR) to (inventory of transuranic actinides in the ALMR at constant power, in the ALMR fuel cycle, and in radioactive waste at time τ).

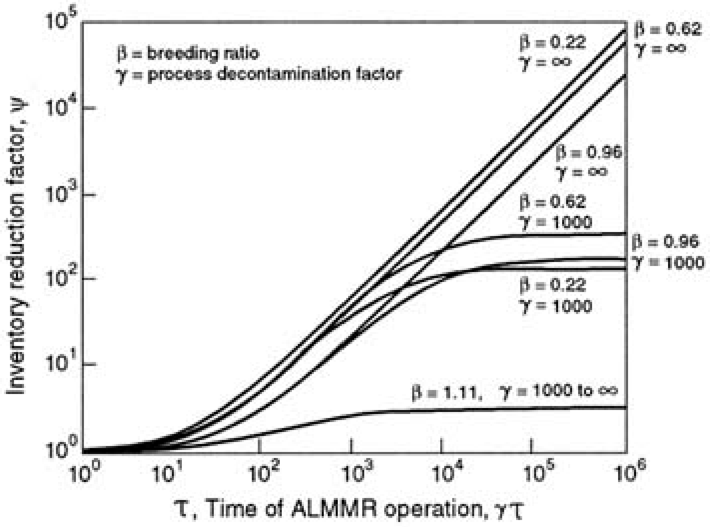

Figure 1 (from Reference 7, Figure 4-9, based on Reference 8)

the fate of this uranium, it appears that it may have to be placed in the repository because of its potential health risk. Because of increased fuel cycle activities, there might also be increases in risk due to potential fuel cycle accidents; we have seen no estimate of this. Proponents of the ALMR think the small unit size of its modular construction makes it safer (due to low coolant pressure and decay heat removal by natural convection) than a conventional light water reactor (Reference 2). If true, this would reduce risk.

What licensing or regulatory problems may be encountered?

Significantly reducing the total inventory of light water reactor-produced actinides by ALMR actinide burners will require an extensive, fully deployed ALMR system. As proposed, such a system would consist of ALMRs with associated ALMR reprocessing facilities, fuel fabrication facilities, and one or more facilities for reprocessing light water reactor spent fuel. In addition, storage and disposal facilities must be provided for the new waste streams, some of which will be high-level waste streams, produced by the ALMR system. Each of these facilities must be licensed.

In the U.S., the Atomic Energy Commission granted operating licenses in the 1960s to one fast breeder reactor, Fermi I, and to the reprocessing plant at West Valley, New York. The licensing processes for any reactor or other nuclear facility are likely to be difficult and contentious; this is especially likely for new types of facilities such as the ALMR and its associated reprocessing plants. Adequate opportunity for U.S. Nuclear Regulatory Commission staff to perform generic rule-making, review standard designs and establish regulatory guides would be mandatory for the ALMR to succeed. Strong local intervention could be expected for each facility.

How would actinide burning affect the need and schedule for the first geological repository?

The statutory limit for the first repository is 70,000 metric tons of uranium until such time as the second repository is in operation. Of this amount, about 63,000 metric tons of uranium is expected to be light water reactor spent fuel, containing about 600 metric tons of transuranic actinides. This corresponds to the amount of spent light water reactor fuel projected to be discharged in the U.S. through the year 2011.

In the actinide burning cycle, a repository would still be required. The repository would receive canisters of high-level waste containing mostly fission products and relatively small quantities of actinide waste from light water reactor fuel and reprocessing of existing defense wastes. Current estimates are that the fission products and remaining actinides would require about the same number of canisters as in the once-through spent fuel cycle (Reference 7, Table 4-5). Hanford defense wastes contain a much smaller amount of transuranics (a total of approximately 500 kg of transuranic actinides); Hanford plans to concentrate and vitrify the fission products and actinides for deep geological disposal. Again, removing the transuranics by transmutation does not appear to reduce the number of waste canisters, although chemical separation can greatly decrease the number of canisters of Hanford defense wastes. The ALMR high-level waste may be in a form different from spent fuel and defense waste (Reference 7); if so, an extensive research program would be required to characterize the ALMR high-level waste before it could be shown to be acceptable for emplacement at the repository.

The primary thermal impact of transuranic actinide removal is the formation of a waste which cools to moderate temperatures after a few hundred years instead of thousands of years. At early times, the thermal output is dominated by Cs-137 and Sr-90 with about 30 yr half-lives. Because of this, for the initially emplaced waste, power density and repository operational heat issues are not changed substantially by actinide removal. The time duration of the thermal excursion and the maximum temperature reached are both substantially reduced by actinide removal. In some scenarios, capacity increases over time could be greater with actinide removal because of reductions in the long-term thermal output. If it is found desirable to keep repository temperatures low, removal of the actinides is beneficial. However, reduced heat generation may be undesirable in a repository in unsaturated rock, such as that at Yucca Mountain, where near-field temperatures above the boiling point of water may be beneficial for an extended time. Additional reduction in the required disposal area could result from removal of cesium and strontium, but separate surface or subsurface storage of these elements would be required for an extended time. Other engineering measures may also substitute for or add to the potential repository area reductions mentioned above.

The current schedule for the first geological repository is determined by actions necessary to characterize the site and submit a license application in 2001 with repository operations beginning in 2010. The schedule for development of the ALMR system, as proposed, indicates that the first reactor and associated ALMR fuel reprocessing plant would provide modified high-level waste around 2020. Achieving this would require the development, design, licensing, and construction of the reprocessing plant and waste fixation plant for light water reactor spent fuel. The ALMR system will directly affect the licensing of the repository in that the resulting waste form must be certified as part of the repository license. This and other related public policy and institutional factors might introduce uncertainty and potential delays into the repository schedule. However, it may be possible, after initial licensing has been obtained and operation of both the repository and the ALMR system have begun, to modify the repository license to accommodate changes in waste form specification and concomitant repository operations. This could allow a decoupling between the licensing schedules of the ALMR and the repository. Thus, the General Electric Company licensing schedule for high-level wastes from the ALMR to the repository may not affect the repository schedule.

However, the ALMR could still affect the repository schedule. The ALMR project expects major commitments to design and construction of reactor and reprocessing plants in 2002-2005 (Reference 2), as early as eight years before the 2010 opening of the repository. Therefore, under this scenario, there would be no reason to send light water reactor spent fuel to the repository; the fuel could be stored at the light water reactor site or at an interim storage facility until needed for ALMRs. Hence, successful development of the actinide-burning ALMR on the planned schedule makes it unnecessary to load commercial high-level waste into the repository until 2020, when the first waste from reprocessing of light water reactor spent fuel for the ALMR would be ready for emplacement (Reference 2).

What are the costs of actinide burning?

The feasibility of an actinide-burning ALMR is strongly affected by the unit cost of reprocessing light water reactor spent fuel. Such cost is uncertain and is likely to be high because of several factors, especially: (1) the low concentration of actinides in light water reactor fuel, (2) the relatively early state of development of the reference pyro-chemical process for light water reactor fuel, (3) the uncertainty of capital costs for all the facilities involved, and (4) the wide variability in the methods and costs of financing an industrial-scale reprocessing facility in the U.S. The large uncertainty about costs in information presented to the Panel has yet to be resolved.

Summary

This report summarizes the Panel's current information concerning actinide burning in ALMRs and the possible repository impacts. Because we are still in the fact-finding stage of our studies, no conclusions have been drawn about this technology; it and other technologies will now be analyzed by the Panel.

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Panel on Separations Technology and Transmutation Systems

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Gregory Choppin, Florida State University
Allen G. Croff, Oak Ridge National Laboratory
Harold K. Forsen, Bechtel National, Inc.
B. John Garrick, PLG, Inc.
John M. Googin, Martin Marietta Energy, Inc.
Hermann A. Grunder, Continuous Electron Beam Accelerator Facility
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Edwin E. Kintner
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Appendix M

Biographies of Committee and Subcommittee Members

+ **JAMES A. BUCKHAM** is president of JAB, Inc., an engineering consulting firm. He specializes in nuclear chemical engineering, specifically in the areas of nuclear fuel reprocessing and nuclear waste management. He previously was president of Allied General Nuclear Services as its last president. He has been an active member of the American Institute of Chemical Engineers, serving as director from 1980-1983. In 1975 he received the Institute's Robert E. Wilson award. He is a member of Sigma Xi, the American Nuclear Society, and the American Chemical Society. He received B.S., M.S., and Ph.D. degrees in chemical engineering from the University of Washington.

◆ **THOMAS A. BURKE** is associate professor of health policy and management at The Johns Hopkins University School of Hygiene and Epidemiology. His work includes the evaluation of population exposure to the environmental pollutants, assessment of environmental risks, and the application of the epidemiology and health risk assessment to public policy. Prior to his appointment at Johns Hopkins, he was deputy commissioner of health for the State of New Jersey. He is a member of the Council of the Society for Risk Analysis and has served on Office of Technology Assessment advisory panels on Risk Assessment of Chemical Carcinogens and Managing Nuclear Materials from Warheads. He is also a member of the National Academy of Sciences committees on Remediation of Buried and Tank Wastes, and Risk Characterization. He received a B.S. from Saint Peter's College, an M.P.H. from the University of Texas, and a Ph.D. in epidemiology from the University of Pennsylvania.

◆ + **GREGORY R. CHOPPIN** is R.O. Lawton Distinguished Professor of Chemistry at Florida State University. During a postdoctoral period at the Lawrence Radiation Laboratory, University of California, Berkeley, he participated in the discovery of mendelevium, element 101. His research and educational activities have been recognized by the American Chemical Society Award in Nuclear Chemistry, the Southern Chemist Award of the American Chemical Society, the Manufacturing Chemist Award in Chemical Education, a Presidential Citation Award of the American Nuclear Society, and honorary D.Sc. degrees from Loyola University and the Chalmers University of Technology (Sweden). His research interest involves the chemistry of the f-Elements, the separation science of the f-Elements, and concentrated electrolyte solutions. He received a B.S. in chemistry from Loyola University, New Orleans, and a Ph.D. from the University of Texas, Austin.

+ **MELVIN S. COOPS** is a retired chemist from the Lawrence Livermore National Laboratory with more than 40 years experience in the chemical processing of radioactive materials. He is an expert in the chemical isolation of the actinide elements, especially plutonium, americium, and curium. His processing experience includes both aqueous separation methods and pyrochemical systems using molten salts and alloys which are utilized for processing radioactive materials with very high thermal-power emissions. He is currently a consultant to Lawrence Livermore National Laboratory, a technical reviewer for the University of Chicago on the Fast Reactor Program, and a technical advisor to Science Applications International Corp. He has a B.S. in chemistry from the University of California, Berkeley, and is a member of the American Nuclear Society.

◆ **ALLEN G. CROFF** is associate director of the Chemical Technology Division at Oak Ridge National Laboratory (ORNL). His areas of focus include initiation and technical management of research and development involving waste management, nuclear fuel cycles, transportation, conservation, and renewable energy. Since joining ORNL in 1974, he has been involved in numerous technical studies that have focused on waste management and nuclear fuel cycles, including: (1) supervising and participating in the updating, maintenance, and implementation of the ORIGEN-2 computer code; (2) developing a risk-based, generally applicable radioactive waste classification system; (3) multidisciplinary development and assessment of actinide partitioning and transmutation; and (4) leading and participating on multidisciplinary national and international technical committees. He has a B.S. in chemical engineering from Michigan State University, a Nuclear Engineer degree from the Massachusetts Institute of Technology, and an M.B.A. from the University of Tennessee.

• **ERSEL A. EVANS** is consultant to the director of Pacific Northwest Laboratory. In prior positions he directed research, engineering, and project management programs for General Electric, Battelle Pacific Northwest Laboratories, Westinghouse, and the U.S. Department of Energy. Nuclear fuels, materials, and facilities were developed and completed for government, industry, and international research, power, fusion, and aerospace programs and nonnuclear projects. He is a member of the National Academy of Engineering and fellow of the American Nuclear Society, the American Society of Metals, the American Ceramic Society, and the American Institute of Chemical Engineers. Awards he has received include those of the American Nuclear Society, Westinghouse, and the National Endowment for the Arts. He received a B.A. from Reed College and a Ph.D. from Oregon State University.

◆ • [1] **HAROLD K. FORSEN** is a senior vice president of Bechtel Corporation. For many years he was responsible for Bechtel's overall corporate technology program and currently is with Bechtel Hanford, Inc., in Richland, Washington, with similar responsibilities. Previously he managed Exxon Nuclear Company's laser isotope separation program (1973-1981), and was professor of nuclear engineering at the University of Wisconsin (1965-1973), where he performed research in plasma physics and fusion reactor engineering. He received B.S. and M.S. degrees from Caltech, and a Ph.D. from the University of California, Berkeley, all in electrical engineering. He is a member of the National Academy of Sciences.

+ **GERHART FRIEDLANDER** is a nuclear chemist, retired, from Brookhaven National Laboratory, where he worked from 1948 to 1992. He is a member of the National Academy of Sciences and the American Academy of Arts and Sciences. His principal research interests are in nuclear reactions and solar neutrino detection. He received a B.S. in chemistry and a Ph.D. in nuclear chemistry from the University of California, Berkeley, and he holds honorary doctorates from Clark University and the University of Mainz (Germany).

◆ **B. JOHN GARRICK** is president and CEO of PLG, Inc., an engineering, applied science, and management consulting firm. His principal accomplishments include his Ph.D. thesis that was the first to advocate quantitative risk assessments for nuclear power plants, building the first team to perform the initial comprehensive and quantitative risk assessment for the commercial nuclear power industry, being a major contributor to the methods employed in risk analysis, and a prime mover in elevating risk assessment to a science and engineering discipline. He is a member of the National Academy of Engineering and a fellow of the American Nuclear Society, the Society for Risk Analysis, and the Institute for the Advancement of Engineering. In 1994 he was appointed to the U.S. Nuclear Regulatory Commission's Advisory Committee on Nuclear Waste. He is past president of the Society for Risk Analysis and has published approximately 200 papers and reports on risk, reliability, engineering, and technology. He received a Ph.D. in engineering and applied science from the University of California, Los Angeles.

◆ + [2] **JOHN M. GOOGIN** was a nuclear physicist and senior staff consultant to the Oak Ridge National Laboratory Y-12 Plant, Martin Marietta Energy Systems, from 1983 to 1994. He received several awards including the Orlando Lawrence Memorial Award, the Fellow of American Society for Metals, the McGraw Hill's Chemical Engineering Award, and the National Academy of Engineering's William J. Kroll Zirkonium Medal. He received a B.S. in physics from Bates College, a Ph.D. in physical chemistry from the University of Tennessee, and an honorary doctorate from Bates College.

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[1] Served as member until August 5, 1993, after which time he served as an unpaid consultant to the committee.

[2] Deceased January 1994.

◆ • **HERMANN A. GRUNDER** is director of the Continuous Electron Beam Accelerator Facility (CEBAF), a U.S. Department of Energy accelerator laboratory for nuclear research. Before coming to CEBAF, he worked at Lawrence Berkeley Laboratory as accelerator group leader for the SuperHILAC-Bevatron/Bevalac Operation and Development, associate director for the Accelerator and Fusion Research Division, and the deputy director of General Sciences. He has also served on a number of committees, including the Nuclear Science Advisory Committee in 1983-1984, High Energy Physics Advisory Panel, Division of Physics of Beams of the American Physical Society, of which he was chair in 1993, and the Los Alamos AT Division Advisory Committee. He also served as subcommittee chair on the BESAC Panel on Neutron Sources in 1992. He received an M.S. in mechanical engineering at Technische Hochschule (Karlsruhe, Germany) and a Ph.D. in experimental nuclear physics from the University of Basel (Switzerland).

◆ **L. CHARLES HEBEL** is manager of technology evaluation, Xerox Palo Alto Research Center, Palo Alto, California. His work focuses on computing sciences and technologies, systems, and the related materials technologies. Previously he served as a research scientist and department head at the Bell Telephone Laboratories and as a research director at Sandia National Laboratories before joining Xerox in 1973. He has served on and chaired numerous committees that evaluated nuclear issues and technologies. From 1977-1979 he represented the United States on the Working Group on Reprocessing, Plutonium Handling and Recycle at the International Nuclear Fuel Cycle Evaluation. He is a fellow of the American Physical Society and has served as chair of the Society's Panel on Public Affairs. He has a Ph.D. in physics from the University of Illinois.

+ **DARLEANE C. HOFFMAN** is Professor of the Graduate School, University of California, Berkeley, and director of the G. T. Seaborg Institute for Transactinium Science, Lawrence Livermore National Laboratory. She is also currently cogroup leader of the Heavy Element Nuclear & Radiochemistry Group, Lawrence Berkeley Laboratory. She is internationally recognized for her research in the field of atom-at-a-time studies of the nuclear and chemical properties of the transfermium elements, spontaneous fission properties, and studies of radionuclide migration in the environment. She received the American Chemical Society's Award for Nuclear Chemistry, the Garvan Medal, and the Iowa State University Alumni Association Distinguished Achievement Award and Citation of Merit. She is a fellow of the American Physical Society and the American Association for the Advancement of Science, and a member of the Norwegian Academy of Sciences and Letters. She received a B.S. and a Ph.D. in physical chemistry from the Iowa State University.

◆ **THOMAS O. HUNTER** is director of the Energy and Environment Program Center at Sandia National Laboratories where he coordinates Sandia's programs in energy systems and environmental management. His current emphasis is on international energy and environment development and supporting information systems. Previously, he was director of Nuclear Waste Management and Transportation at Sandia; manager of the Yucca Mountain Project; and leader of the Research and Development Program for the Waste Isolation Pilot Plant. He was also responsible for the development of technology for advanced conception in underground nuclear weapons testing, reactor safety programs, and fusion engineering. He received a B.S.M.E. from the University of Florida, an M.S.M.E. from the University of New Mexico, an M.S.N.E. and Ph.D. in nuclear engineering from the University of Wisconsin.

• **WILLIAM M. JACOBI** is a consultant in nuclear technology. He specializes in nuclear system design, nuclear fuel design, radioactive waste management, and nuclear operations. He recently retired from the Westinghouse Electric Corporation, where over a period of years he served as general manager of the Nuclear Technology Division, of the Nuclear Fuel Division, and of the Advanced Power Systems Business Unit. He also served as president of the Westinghouse Hanford Company, and vice president of Government Operations. He received a B.Ch.E. from Syracuse University, an M.Ch.E. from the University of Delaware, and a Ph.D. from Syracuse University.

◆ • **MUJID S. KAZIMI** is professor and department head of the Department of Nuclear Engineering, Massachusetts Institute of Technology. His principal fields of interest are thermal and safety analysis of nuclear systems, including power reactors, fusion devices, and waste storage facilities. He is a member of the American Nuclear Society, the American Society of Mechanical Engineers, and the American Institute of Chemical Engineers. He received a B.Eng. in nuclear engineering from the University of Alexandria (Egypt) and an M.S. and Ph.D. in nuclear engineering from the Massachusetts Institute of Technology.

+ [3] **C. JUDSON KING** is vice provost for research of the University of California system and professor of chemical engineering at the Berkeley campus. He was formerly provost, Professional Schools and Colleges; dean of the College of Chemistry; and chair of the Department of Chemical Engineering, all at the Berkeley campus. He is a member of the National Academy of Engineering. He has received the William H. Walker, Warren K. Lewis, and Clarence G. Gerhold Awards of the American Institute of Chemical Engineers; the Mac Pruitt Award of the Council for Chemical Research; and the George Westinghouse Award of the American Society for Engineering Education. His research interests center around separation processes, presently spray drying, and the use of reversible chemical complexation for recovery of polar organic (carboxylic acid, Glycols, etc.) from aqueous solution. He received a B.E. from Yale University and an S.M. and a Sc.D. in chemical engineering from the Massachusetts Institute of Technology.

◆ • **EDWIN E. KINTNER**, chair of the Subcommittee on Transmutation, was executive vice president of GPU (General Public Utilities) Nuclear Corporation and member of its Board of Directors until his retirement in June 1990. His background includes many years of nuclear reactor experience both with the military and private industry. He was chair of the Nuclear Power Division Advisory Committee and chair of the Advanced Light Water Reactor Utility Steering Committee, which is reconceptualizing the next generation of reactors. In addition, he was a member of the M.I.T. Corporation Visiting Committee for Nuclear Engineering. He is a member of the National Academy of Engineering and the American Nuclear Society. Prior to joining GPU, he was director of the U.S. Magnetic Fusion Program at the U.S. Department of Energy and its predecessor agency. In previous years, he was assistant director for reactor engineering and then deputy director of the former Atomic Energy Commission's Reactor Development Division. Among awards received during his career are the Secretary of the Navy Commendation Medal, the title of Distinguished Alumnus of the Massachusetts Institute of Technology, and the highest award of the Senior Executive Service. He represented United States as chair of the U.S./U.S.S.R. Joint Fusion Power Coordinating Committee and chair of the U.S/Government of Japan Fusion Power Coordinating Committee. He was the U.S. representative to the International Fusion Research Council, to the International Energy Agency's Fusion Power Coordinating Committee, and chair of the U.S. Fusion Power Coordinating Committee. He received a B.S. from the U.S. Naval Academy and an M.S. in nuclear physics and an M.S. marine engineering, both graduate degrees from the Massachusetts Institute of Technology.

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[3] Resigned as a member of the Subcommittee on Separations as of March 1994.

◆ + **ROLLAND A. LANGLEY** is the president and CEO of BNFL Inc., the American subsidiary of British Nuclear Fuels plc. His extensive nuclear industry experience, including 27 years with Bechtel, covers the full range of nuclear fuel cycle and nuclear decontamination activities, as well as fast breeder reactor and light-water reactor plant designs. A graduate of the University of California at Berkeley in engineering physics, he also received an M.S. in mechanical engineering from the University of Pittsburgh and an M.B.A. in business management from Golden Gate University.

• **JOHN C. LEE** is a professor of nuclear engineering at the University of Michigan. He is well recognized for his contributions in nuclear reactor physics, reactor safety analysis, a power plant simulation and control. He is a fellow of the American Nuclear Society and has received the Distinguished Service Award from the University of Michigan Class of 1938E. He received a B.S. from Seoul National University (Korea) and a Ph.D. from the University of California, Berkeley.

• **GLENN E. LUCAS** is professor and vice chair of the Department of Chemical and Nuclear Engineering at the University of California, Santa Barbara. He concurrently holds part-time appointments in the departments of Materials and Mechanical Engineering. His teaching and research interests lie in the area of structural materials for advanced engineering and aerospace applications and the effects of service conditions on mechanical property degradation, topics on which he has authored or coauthored over 90 publications. He has received numerous awards, including the American Nuclear Society's Young Member Engineering Achievement Award. He has participated in a variety of advisory committees to government and industry and was chair of the Materials Science and Technology Division of the American Nuclear Society. He received a B.S. from the University of California, Santa Barbara, and a S.M. and Sc.D. from the Massachusetts Institute of Technology.

◆ **EDWARD A. MASON** is a consulting chemical and nuclear. He specializes in nuclear reactor safety, nuclear chemical engineering, and technology development and management. Previously he was director of research at Ionics, Incorporated; associate professor of chemical engineering, professor and head of nuclear engineering at the Massachusetts Institute of Technology; a commissioner of the U.S. Nuclear Regulatory Commission; and vice president of research at Amoco Corporation. He is a member of the National Academy of Engineering (where he also served on the Council), and he serves on the boards of directors of Unicom and Symbollon. He is a fellow of the American Academy of Arts and Sciences, the American Institute of Chemical Engineers, the American Nuclear Society, the American Association for the Advancement of Science, and the New York Academy of Sciences. He received a B.S. in chemical engineering from the University of Rochester, a S.M. and Sc.D. also in chemical engineering from the Massachusetts Institute of Technology.

◆ + **FRED W. MCLAFFERTY**, chair of the Subcommittee on Separations, is professor of chemistry at Cornell University. He is an analytical chemist specializing in molecular mass spectrometry, including its coupling with separation devices. After combat infantry service in World War II, he received an M.S. from the University of Nebraska and a Ph.D. from Cornell. He spent 14 years at the Dow Chemical Company, 8 of those years as director of the Eastern Research Laboratory, and 4 years at Purdue University. He has served on five National Research Council boards and panels and has been a member of the National Academy of Sciences since 1982.

+ **ROBERT A. OSTERYOUNG** is professor and head of the Department of Chemistry, North Carolina State University. His major research interests are in molten salt chemistry, electrochemistry, and electroanalytical chemistry, particularly in pulse voltammetric methods. He has received several awards for his work including the American Chemical Society Division of Analytical Chemistry Award in Electrochemistry and the Electrochemical Society's Physical Electrochemistry Division Max Bredig Award in Molten Salt Chemistry. He is a fellow of the American Association for the Advancement of Science and the Electrochemical Society. He received a B.S. in chemistry from Ohio University and an M.S. and Ph.D. from the University of Illinois.

◆ • **THOMAS H. PIGFORD** is professor of nuclear engineering at the University of California, Berkeley. He is an international consultant in the geologic disposal of radioactive waste. He specializes in the nuclear fuel cycle, nuclear safety, environmental analysis of nuclear systems, and prediction of the release of radionuclides from buried solid waste and their transport through geologic media. He has received many awards for his achievements in engineering, including the Robert E. Wilson Award and the Service to Society Award from the American Institute of Chemical Engineers, the Arthur H. Compton Award from the American Nuclear Society, and the John Wesley Powell Award from the U.S. Geological Survey. He is a member of the National Academy of Engineering and has served on many of its panels and boards. He was a member of the Presidential Commission on the Accident at Three Mile Island. He earned a B.S. from the Georgia Institute of Technology and a S.M. and Sc.D. in chemical engineering from the Massachusetts Institute of Technology.

◆ **NORMAN C. RASMUSSEN**, chair of the Committee on Separations Technology and Transmutation Systems, is McAfee Professor of Engineering, emeritus and a senior lecturer at the Massachusetts Institute of Technology. He specializes in risk assessment methodologies for complex engineered systems. Most of his work has been on the risk assessment of nuclear power reactors. Since his retirement in 1994, he continues to work at MIT on a part-time basis. He served as director of the Reactor Safety Study (WASH 1400) for the Nuclear Regulatory Commission. He is a member of the National Academy of Engineering and the National Academy of Sciences. He received numerous awards for his contribution to nuclear reactor safety, including the Enrico Fermi Award from the U.S. Department of Energy in 1986. He received a B.A. from Gettysburg College and a Ph.D. in physics from the Massachusetts Institute of Technology.

• **KUNIHIKO UEMATSU** is director-general of the Organization for Economic Cooperation and Development Nuclear Energy Agency, based in Paris, France. He is a highly renowned expert in the field of nuclear engineering, and was previously executive director of the Power Reactor & Nuclear Fuel Development Corporation of Japan. He has also taught at several major Japanese universities and is the author of many publications on nuclear engineering. He is a member of the American Nuclear Society and other prestigious societies. After graduating from Kyoto University, and completing graduate school at the University's School of Engineering, he received a Sc.D. from the Department of Nuclear Engineering at the Massachusetts Institute of Technology.

◆ [4] **DAN W. REICHER** is deputy chief of staff and environmental counsellor in the Office of the Secretary of the U.S. Department of Energy, and is adjunct professor of nuclear regulations at the Maryland Law School. From 1983-1993 he was a senior attorney at the Natural Resources Defense Council where he directed both national and international defense and environmental projects. His special interests include environmental law, regulations, and standards. Over the years he has served on several advisory groups, including for the U.S. Congress, the Office of Technology Assessment, the Review Panel on Managing Low-Level Radioactive Waste, the Keystone Center, and the Department of Defense Hazardous Waste Management. He has been a member of the Board of Directors of the Americans for the Environment since 1986. He received a J.D. from Stanford University Law School.

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[4] Resigned June 1, 1993.

◆ **JAMES E. WATSON, JR.** is a professor in the Department of Environmental Sciences and Engineering at the University of North Carolina. He directs a graduate education program in radiation protection and teaches and conducts research in that program. Previously he was president of the Health Physics Society and chair of the Radiological Health Section of the American Public Health Association. He was selected as a National Lecturer for Sigma Xi, and has served on the U.S. Environmental Protection Agency's Science Advisory Board and the North Carolina Radiation Protection Commission. He received a B.S. in nuclear engineering, an M.S. in physics from North Carolina State University, and a Ph.D. in environmental sciences and engineering from the University of North Carolina.

◆ **SUSAN D. WILTSHIRE** is vice president at JK Research Associates, a consulting firm specializing in public policy formulation of environmental and community planning and public involvement in planning for technical programs. She is particularly involved in issues regarding radioactive waste management and environmental restoration. She serves on advisory committees to the U.S. Environmental Protection Agency and the National Council on Radiation Protection and Measurements. She is also a member of the National Research Council's Committee on the Technical Bases for Yucca Mountain Standards and the Committee to Review New York State's Siting and Methodology Selection for Low Level Radioactive Waste Disposal, which she also chairs. She is chair of the Board of Cape Ann and Northeast Health Systems and previously served as chair of the elected Board of Selectmen of Hamilton. She is a former president of the League of Women Voters of Massachusetts and is author of the current edition of the U.S. League of Women Voters' *A Nuclear Waste Primer*. She received a B.Sc. from the University of Florida.

◆ [5] **RAYMOND G. WYMER** is a retired director of the Chemical Technology Division of Oak Ridge National Laboratory. He is a specialist in radiochemical separations technology for radioactive waste management and nuclear fuel reprocessing. He is a consultant for the Oak Ridge National Laboratory and for the U.S. Department of Energy in the area of chemical separations technology. He consults for the U.S. Department of State and the U.S. Department of Energy on matters of nuclear nonproliferation. He is a fellow of the American Nuclear Society and the American Institute of Chemists, and has received the American Institute of Chemical Engineers Robert E. Wilson Award in Nuclear Chemical Engineering and the American Nuclear Society's Special Award for Outstanding Work on the Nuclear Fuel Cycle. He received a B.A. from Memphis State University and an M.A. and Ph.D. from Vanderbilt University.

+ Separation Subcommittee Member
◆ STATS Main Committee Member
• Transmutation Subcommittee Member Integration Subcommittee

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[5] Resigned September 30, 1993.

Appendix N

Glossary of Terms

absorption	Incorporation of the physical or molecular structure (see *sorption*).
accessible environment	Those portions of the environment directly in contact with or readily available for use by human beings. Includes the Earth's atmosphere, the land surface, aquifers, surface waters, and part of the oceans.
actinide	An element with an atomic number from 89 to 103, inclusive. All are radioactive.
activation product	An element made radioactive by bombardment with neutrons.
activity	For an amount of radioactive nuclide in a particular energy state at a given time, the quotient of dN by dt is the expectation value of the number of spontaneous nuclear transitions from that energy state in the time interval dt. The SI unit of activity is the becquerel (Bq); the curie (Ci) is being phased out.
Alara	An acronym for "as low as reasonably achievable" a concept meaning that the design and use of sources, and the practices associated therewith, including their disposal, should be such as to ensure that exposures are kept as low as is reasonably practicable, economic and social factors being taken into account.
alpha-bearing waste	Waste containing one or more alpha emitting radionuclides, usually actinides, in quantities above acceptable limits for uncontrolled release. The limits are established by the national regulatory body.
aluminosilicate glass	A durable type of glass in which some of the silicon atoms that normally form the amorphous, polymeric structures that are characteristic of glasses (networks) are replaced by aluminium atoms. Aluminosilicate glasses are candidate matrices for solidifying some kinds of radioactive waste.

annual dose equivalent limit	The value of the annual dose equivalent that must not be exceeded, according to the International Commission on Radiological Protection (ICRP) system of dose limitation.
anoxic condition	A chemical condition, often existing in underground waste repositories, in which the partial pressure of oxygen in the groundwater is very low. This affects the oxidation state of chemical species in the groundwater as well as bacteriological processes that can occur.
aquifer	A water-bearing formation below the surface of the earth. An aquifer can furnish an appreciable supply of water for a well or spring.
area survey	The first stage of siting a waste repository, during which a broad area is examined to eliminate obviously unsuitable regions and to identify other regions that may contain suitable sites.
argillaceous	A term applied to all rocks and substances composed of clay or having a notable proportion of clay in their composition.
arid sites	A term often applied to a shallow, land waste-disposal site located in an area that receives very little annual precipitation, typically less than 250 mm per year.
back end of the fuel cycle	The part of the fuel cycle that includes spent-fuel storage, fuel reprocessing, mixed-oxide fuel fabrication, and waste management including spent-fuel disposal.
backfill	The material used to refill the excavated portions of a repository or of a borehole after waste has been emplaced.
barrier (natural or engineered)	A feature that delays or prevents material migration to or from disposal components. Facilities may include multiple barriers.
becquerel (Bq)	The SI unit of radioactivity, equivalent to 1 disintegration per second (approximately 2.7×10^{-11} Ci).

bedded salt	A salt formation in which the salt is roughly horizontal, laterally extensive, and relatively thin in the vertical direction.
bentonite	A soft, plastic light-colored clay formed by chemical alteration of volcanic ash. It is composed essentially of montmorillonite and related minerals of the smectite group. The properties of bentonite depend largely on its ion exchange characteristics. Bentonite may be used as a buffer material for surrounding waste packages in a deep repository.
biosphere	That portion of the Earth's environment inhabited by any living organisms. It comprises parts of the atmosphere, the hydrosphere (oceans, seas, inland waters and subterranean waters), and the lithosphere. The biosphere includes the human habitat or environment in the widest sense of these terms.
bitumen	A general name for various solid and semisolid hydrocarbons that are fusible and are soluble in carbon bisulphide. Petroleums, asphalts, natural mineral waxes, and asphaltites are all considered bitumens. Bitumen is known to be very stable in the terrestrial environment and is sometimes used as a matrix for immobilizing low-and intermediate-level waste.
borehole	A cylindrical excavation, made by a rotary drilling device. For disposal at relatively shallow depth, boreholes can be drilled from the surface; for deep disposals, they can be drilled from an access shaft in a mine or from the surface.
borosilicate glass	(1) A supercooled liquid based on a random lattice of silica tetrahedra, modified with boron and other cations. (2) A glass composition used as an immobilization matrix for a radioactive waste.
brine	An aqueous solution containing a high concentration of dissolved salts. Any water in a repository in a salt formation, for example, would certainly be brine, and its ability to corrode waste forms would be expected to differ considerably from ordinary groundwater.
buffer material	Any substance, frequently a natural clay, placed around a waste container in a repository. Often, a primary purpose of such material is to serve as an additional barrier to prevent water from contacting the waste container and, by adsorption, to reduce the rate of radionuclide migration from the waste into the repository.

burial ground	An area of land that has been dedicated for the shallow disposal of short-lived low-and medium-level waste. Access by the public, as well as future use of the land, will probably be restricted.
canister (can)	A closed or sealed container for nuclear fuel or other radioactive material, which isolates and contains the contents; it may rely on other containers (e.g., a cask) for shielding.
cask	A massive container to transport or store irradiated nuclear fuel and other radioactive materials. It provides chemical, nuclear, and radiological protection and dissipates heat generated by radioactive decay.
cement	A standard material used by the construction industry that has many uses in waste management because of its low cost and ease of handling. Properties of cement mixtures and of the final solid product can be modified considerably by the use of additives. The mixture of final product is referred to as "concrete" if it contains aggregate (usually small stones) or without aggregate, "a grout."
cementation	The process of incorporating wastes into a concrete matrix as a means of immobilization.
ceramic materials	Solid materials, usually containing SiO_2 and metal oxides, that generally require fabrication at an elevated temperature (typically greater than 800° C) and often elevated pressure. Their microscopic structure is crystalline, which distinguishes them from amorphous glasses. They are very stable and have been considered as good candidates for solidifying high-level waste.
cladding (material)	An external layer of material (usually of Zircaloy, stainless steel, magnesium) directly surrounding nuclear fuel or other substance that seals and protects it from the environment and protects the environment from radioactive material produced during irradiation. For high temperature reactor (HTR) fuel particles, the multilayer protective claddings are known as coatings.

clay	Minerals that are essentially hydrous aluminium silicates, and occasionally hydrous magnesium silicates, with sodium, calcium, potassium and magnesium cations. Also denotes a natural material with plastic properties, which is essentially a composition of fine to very fine clay particles. Clays differ greatly mineralogically and chemically and consequently in their physical properties; especially because of their large surface areas, most of them have good sorption characteristics.
cold testing	Testing of method, process, apparatus, or instrumentation with the highly radioactive materials replaced by nonradioactive materials or materials that may contain radioactive tracers.
competent authority	An authority designated or otherwise recognized by a government for specific purposes in connection with radiation protection or nuclear safety.
complexation	A chemical term that refers to complex formation, in which a central metal ion is chemically bonded to atoms surrounding it. Depending on the circumstances, complexation can be used to remove radionuclides from solution, or by the formation of a neutral molecule, it may greatly enhance the mobility of a radionuclide in the environment.
computer code	See *computer model*.
computer model	A mathematical description of a facility or sequence of events that is evaluated via a computer. Computer models are usually indispensable for performing a safety analysis of a waste facility. In particular, models are used extensively to evaluate long-term events associated with a waste repository that cannot be tested directly and to predict the resulting radiation dose to man.
conditioning of waste	Those operations that transform waste into a form suitable for transport or storage or disposal. The operations may include converting the waste to another form, enclosing the waste in containers, and providing additional packaging.
confinement (or isolation) of waste	The segregation of radionuclides from the human environment and the prevention of their release into that environment in unacceptable quantities or concentrations.

containment	A term signifying either: (1) the confinement of radioactive material in such a way that it is prevented from being dispersed into the environment or is only released at a specified rate or (2) the device used to effect such confinement.
contamination, radioactive	The presence of a radioactive substance or substances in or on a material or in a place where they are undesirable or could be harmful.
controlled area	An area where workers might receive doses in excess of three-tenths of the occupational dose equivalent limits during the anticipated working period and where appropriate controls (such as restricted access, individual assessment of dose, and special health supervision) are accordingly applied.
critical group	For a given radiation source, the members of the public whose exposure is reasonably homogeneous and is typical of individuals receiving the highest effective dose equivalent or dose equivalent (whichever is relevant) from the source.
crystalline rock	(1) A rock consisting of minerals in an obviously crystalline state. (2) An inexact general term for igneous and metamorphic rocks as opposed to sedimentary ones (see *granite*).
curie (Ci)	The former unit of activity, equal to 3.7×10^{10} becquerels.
decommissioning	The work required for the planned permanent retirement of a nuclear facility from active service. Different regulations will apply thereafter.
decontamination	The removal of radioactive contaminants with the objective of reducing the residual radioactivity level in or on materials, persons, or the environment.
deep geological repository	A repository constructed, usually in consolidated rock, at a depth of several hundred meters or more in a continental formation.

de minimis	Part of the maxim *de minimis non jurat lex* (the law does not concern itself with trifles), sometimes used with reference to sources of radiation that a competent authority may decide to exempt from defined regulatory requirements, because individual and collective effective dose equivalents received from them are both so low that they may be ignored.
dispersion	The summed effect of the processes of transport, diffusion, and mixing, which tend to distribute materials from wastes or effluents through an increasing volume of water or air. The ultimate effect appears as a dilution of the materials.
disposal	The emplacement of waste in a repository, or at a given location, without the intention of retrieval. Disposal also covers the approved direct discharge of wastes into the environment, with subsequent dispersion.
disruptive event	An event (e.g., faulting, meteorite impact) that disrupts a waste repository.
distribution coefficient	A quantitative measure of how a given chemical species partitions itself between two phases at equilibrium. In waste processing, this parameter is used to predict the effectiveness of separation methods such as solvent extraction, ion exchange, or gas scrubbing. In environmental studies, the quantity is sometimes used to predict how soils or backfill materials can retard radionuclide movement.
documentation	Written, recorded, or pictorial information describing, defining, specifying, reporting, or certifying activities, requirements, procedures, or results.
domal (or dome) salt	A local geologic formation of salt in which the salt thickness is greater vertically than laterally. The top of the formation may bear resemblance to a dome or to a mushroom.
dose assessment	An estimate of the radiation exposure of an individual or population group, usually by means of predictive modeling techniques, sometimes supplemented by the results of measurements.
emanation	Radioactive gas formed by decay of a radioactive solid. The emanation may or may not be retained within the pore space of the solid phase.

embedding	A process of putting solid or liquid waste into a matrix to form a heterogeneous waste form.
emplacement	Placing the waste in its location for storage or disposal.
emplacement density	Amount of waste emplaced per unit area or volume of a storage or disposal site (e.g., canisters per hectare).
engineered storage	The storage of radioactive wastes, usually in suitably sealed containers, in any of a variety of structures especially designed to protect them and to help keep them from leaking to the biosphere by accident or sabotage. They may also provide for extracting heat of radioactive decay from the waste.
entombment decommissioning	Placement of radioactive wastes and structural materials within an entombment structure (often comprising a portion of the existing production structure) for permanent disposal. Only those materials with hazardous lifetimes, as determined by radiological assessments, less than or equal to the expected lifetime of the entombment structure are intended to be so placed. Other radioactive materials are removed from the site for disposal.
environmental restoration project	A group of activities initiated to access a Department of Energy facility or radioactive waste site that may require restoration to acceptable radiation levels.
exclusion area	A term used in some countries to designate a zone that may be established around a nuclear facility or other radiation source, to which access is permitted under controlled conditions and in which residence is normally prohibited.
exposure	Irradiation of persons or materials. Exposure of persons to ionizing radiation may be either: 1. external exposure (irradiation by sources outside the body) or 2. internal exposure (irradiation by sources inside the body). The term occupational exposure refers to exposure of a worker received or committed during a period of work.

far-field	Rock formations outside of the repository, including the surrounding strata, at a distance from the waste disposal site such that, for modeling purposes, the site may be considered as a single entity, and the effects of individual waste packages are indistinguishable in the effects of the whole.
fission product	A nuclide produced either by fission or by the radioactive decay of nuclides formed by fission.
fissure	An extensive crack, break, or fracture in the rock.
fixation (of radionuclides)	The practice of immobilizing radionuclides so that they are not easily dispersed. The term often refers to the application of paint or a similar material to a contaminated surface in order to prevent the radionuclides from becoming airborne or transferred by casual contact.
food chain (or web)	A figure of speech for the dependence of organisms on others for food in a series, beginning with plants or scavenging organisms and ending with the largest carnivores. A web is a network or series of food-chains.
formerly utilized site	A site contaminated with radioactive wastes that was previously used for supporting nuclear activities of the Department of Energy's predecessor agencies, the Manhattan Engineering District (Manhattan Project) and the Atomic Energy Commission.
fracture	A crack, joint, fault, or other break in rock. In underground repositories, fractures are of concern as possible paths for water flow and radionuclide migration.
front end of the fuel cycle	Mining, milling, enrichment, and fabrication of nuclear fuel; sometimes irradiation in reactors is included.
fuel reprocessing plant	Plant where spent-fuel elements are dissolved, waste materials are removed, and reusable materials are segregated.

geological repository	A final disposal facility located deep underground in a stable formation such as salt, granite, etc. Usually such a repository would be provided for long-lived radioactive waste such as alpha bearing or high-level waste or spent fuel.
glass ceramic	The product resulting after a glass has been transformed into a crystalline material by a controlled process such as heating. The product may retain the desirable properties of both a glass and a ceramic (see *ceramic materials*).
granite	Broadly applied, any holocrystalline quartz-bearing plutonic rock. Granite formations are being considered as possible hosts for repositories deep underground for high-level waste.
gray (Gy)	The SI unit of absorbed dose equal, for ionizing radiation, to 1 joule of radiant energy absorbed in 1 kg of the material of interest ($1\ \text{Gy} = 1\ \text{J}\bullet\text{kg}^{-1}$).
half-life	In physics, the time required for the transformation of one-half of the atoms in a given radioactive decay process, following the exponential law (physical half-life). By analogy, in biology this term is used in connection with the clearance of a substance from a tissue, an organ, or the whole body (when the kinetics of such a phenomenon roughly follow an exponential dependence) to mean the time for one-half of this substance to be eliminated (biological half-life). The time necessary for a radioactive material in a living organism to be reduced to one-half of its initial value by a combination of biological elimination and radioactive decay is termed effective half-life.
high-level waste (HLW)	(1) The highly radioactive liquid, containing mainly fission products, as well as some actinides, which is separated during chemical reprocessing of irradiated fuel (aqueous) waste from the first solvent extraction cycle and those waste streams combined with it. (2) Spent reactor fuel, if it is declared a waste. (3) Any other waste with a radioactivity level comparable to (1) or (2).
host rock (or host medium)	A geological formation in which a repository is located.
hulls and spacers	Radioactive waste, comprising cladding hulls and assembly grid spacers, generated during reprocessing when spent-fuel assemblies are disassembled and the fuel is dissolved.

humid sites	A near-surface repository located in an area at which annual precipitation exceeds water loss by evaporation, hence there is a significant downward flux of moisture through the soil that could transport radionuclides. Uptake of radionuclides by plant roots may also be significant in a humid site.
hydrogeology	The study of the geological factors related to the Earth's water.
hydrology	The study of all waters in and on the Earth. It includes underground water, surface water, and rainfall and embraces the concept of the hydrological cycle.
ingest	Take into the body by way of the digestive tract.
institutional control	Control by an authority or institution designated under the laws of a country or state. This control may be active (monitoring, surveillance, remedial work) or passive (land-use control).
interim storage (storage)	Storage of radioactive materials such that:
intermediate-level waste; medium-level waste	Waste of a lower activity level and heat output than high-level waste, but which still requires shielding during handling and transportation. The term is used generally to refer to all wastes not defined as either high-level or low-level (see *alpha-bearing waste* and *long-lived waste* for other possible limitations).
International Commission on Radiological Protection (ICRP) limit	A primary dose equivalent limit recommended by the ICRP. Dosimetric models may be used to derive the annual limit on intake and derived air concentration.
intrusive rock	A body of igneous rock that has forced itself into an existing rock formation.
ion exchange	A usually reversible exchange of one ion with another, either in a liquid, on a solid surface, or within a crystalline lattice.

isolation of waste	See *confinement of waste*.
joule melters	An electrically powered glass-making furnace in which the molten glass itself carries the electric current and is thereby heated. Such a design is considered to be well suited for vitrifying radioactive waste.
leachability	The susceptibility of a solid material to having its soluble, sorbed, or suspendable constituents removed by the dissolving or erosive action of water or other fluids.
leachate	A solution, typically groundwater, that has been in contact with radioactive waste and as a result may contain radionuclides.
leaching	(1) Extraction of a soluble substance from a solid by a solvent with which the solid is in contact. (2) The term is often used in waste management to describe the gradual dissolution/erosion of emplaced solid waste or chemicals therefrom, or the removal of sorbed material from the surface of a solid or porous bed.
leach rate	The rate of dissolution or erosion of material from a solid. The term may be used to describe the rate of gradual dissolution/erosion of emplaced solid waste or the removal of sorbed material from the surface of a solid or porous bed.
leach test	A laboratory test conducted to determine the rate at which radionuclides are released from a waste form that is in contact with water. These tests are considered to be essential for judging and comparing waste forms. Many different test parameters have been used, and a number of protocols have been published.
lithostatic pressure	Pressure underground due to the weight of overlying rock, soil, or water.
long-lived waste	Waste that will not decay to an acceptable activity level in a period of time during which administrative controls can be expected to last (see *short lived waste*).

low-level waste (LLW)	Waste that, because of its low radionuclide content, does not require shielding during normal handling and transportation (see *alpha-bearing waste* and *long-lived waste* for other possible limitations).
lysimeter	A device that provides containment for conducting migration experiments under ambient outdoor conditions. A typical lysimeter could be a large-diameter (2 m or more) pipe emplaced vertically in the ground, with its open end several centimeters above the surface and its lower end sealed. Rain water percolating through a mixture of waste and soil would reach the closed end, where it would be pumped back to the surface for analysis.
magnox	A magnesium alloy used as the cladding material in some gas-cooled reactors. Such reactors are known as "Magnox reactors."
matrix	In waste management, a nonradioactive material used to immobilize radioactive waste in a monolithic structure. Examples of matrices are bitumen, cement, various polymers, etc.
maximum permissible concentration (MPC)	Maximum levels of radioactivity in drinking water or in air as established by national authorities.
mixed waste	Radioactive waste that also contains chemicals that could cause undesirable effects in the environment. Such wastes present a number of technical and regulatory problems as regards processing and disposal.
model	In applied mathematics, an analytical or mathematical representation or quantification of a real system and the ways that phenomena occur within that system. Individual or subsystem models can be combined to give system models. Deterministic and probabilistic models are two types of mathematical models.
monitoring	The methodology and practice of measuring levels of radioactivity either in environmental samples or en route to the environment. Examples include groundwater monitoring, gaseous effluent (stack) monitoring, and personnel monitoring.

multibarrier	A system using two or more independent barriers to isolate the waste from the human environment. These can include the waste form, the container (canister), other engineered barriers, and the emplacement medium and its environment (see *barrier*).
natural analogues	Radioactive minerals or mineral deposits, or even man-made objects, whose migration history or corrosions over very long times can be determined and used to forecast the possible behavior of chemically similar waste radionuclides.
off-gas	The gas streams that arise from a process. Typical processes in radioactive waste management facilities, such as dissolution, evaporation, incineration, vitrification, bituminization, and cementation, will generate process off-gases that contain water vapor, acid vapors, aerosols, radioactive constituents, and gaseous chemical constituents.
off-gas treatment	The removal of radioactive components or chemical pollutants from gases prior to their release under controlled conditions into the atmosphere.
operating records	A set of documents, such as instrument charts, certificates, log books, computer printouts, and magnetic tapes, kept at each nuclear facility and organized in such a way that they provide a complete and objective history of the operation of the facility.
operational period	The period during which a nuclear facility is being used for its intended purpose after which it is shut down and decommissioned.
operations, waste management	Broad classification of waste management activities in terms of their basic function (e.g., waste storage, treatment, transportation, or disposal).
operator	Any person, government, or other entity that conducts or carries on operations at a nuclear facility.
overpack	Secondary (or additional) external containment for packaged radioactive waste.

pathways model	A mathematical description, usually in the form of a computer algorithm, that determines the relative significance of possible radionuclide transport vectors, (e.g., air, groundwater, surface water, intrusive roots, animals, etc.).
percolating groundwater	Groundwater that seeps via saturated flow conditions through soil or rock strata (see *saturated zone*).
performance assessment	Analysis to predict the performance of the system or its subsystem, followed by comparison of the results of such analysis with appropriate standards or criteria. When the system under consideration is the overall waste disposal system and the performance measure is radiological impact or some other global measure of impact safety, performance assessment becomes the same as safety assessment.
permeability (of rock)	The capacity of a porous or previous rock to transmit a fluid (see *darcy*).
plasticity	The property of a material (e.g., rock salt) that enables it to undergo permanent deformation without appreciable volume change or elastic rebound and without rupture.
porosity	The ratio of the aggregate volume of interstices in a rock or soil to its total volume.
porous media	Material that contains pores or cracks through which water or gas can flow. Often the term is applied to the geological formations around a waste repository and could denote an undesirable situation.
postsealing period	The period after a waste repository has been shut down and sealed.
preliminary site selection	The second stage of siting a waste repository, during which a manageable number of potentially suitable sites is examined, using existing information or information from limited explorations, to determine whether one or more regions contain sites suitable for additional site confirmation studies.

pretreatment of waste	Any step carried out prior to operations that have been defined as treatment, conditioning, offsite transport, or disposal. Pretreatment techniques and practices include: —collection and segregation, —size reduction, and —chemical adjustment.
primary waste	As-generated form and quantity of a waste.
quality assurance	Planned and systematic actions aimed at providing adequate confidence that an item of a facility will perform satisfactorily in service.
quality control	Actions that provide a means to control and measure the characteristics of an item, process, facility, or person in accordance with quality assurance requirements.
rad	A unit of absorbed dose of ionizing radiation equal to one-hundredth of a gray (1 cGy).
radiation damage	Deleterious changes in the physical or chemical properties of a material resulting from exposure to ionizing radiation. This term does not apply to biological systems.
radiation protection or radiological protection	(1) All measures associated with the limitation of the harmful effects of ionizing radiation on people, such as limitation of external exposure to such radiation, limitation of bodily incorporation of radionuclides, and the prophylactic limitation of bodily injury resulting from either of these. (2) All measures designed to limit radiation-induced chemical and physical damage in materials.
radioactive material	A material of which one or more constituents exhibit radioactivity.
radioactive source term	An expression used to denote information about the actual or potential release of radioactive material from a given source, which may include a specification of the composition, the amount, the rate, and the mode of the release.

radioactive waste management	All activities, administrative and operational, that are involved in the handling, treatment, conditioning, transportation, storage, and disposal of waste.
radiolysis	Chemical decomposition caused by ionizing radiation.
radiolytic effect	An effect caused by radiation-induced degradation of chemical compounds.
radionuclide migration	The movement of radionuclides through various media due to fluid flow or by diffusion.
radionuclide transport	The action of a particular vector that results in movement of radionuclides in the environment (e.g., radionuclide transport by ground water). This specific term does not refer to the intentional transportation of radioactive materials by man (e.g., transportation of nuclear wastes in transport casks, etc.).
reactor waste	Waste arising from the routine operation of a nuclear reactor.
reinserted fuel	Irradiated reactor fuel that is discharged in one cycle and inserted in the same reactor during a subsequent refueling. In a few cases, fuel discharged from one reactor has been used to fuel a different reactor.
rem	A unit of dose equivalent to one-hundredth of a sievert (Sv).
remedial action	Corrective measures imposed at a near-surface repository to prevent a previously unforeseen circumstance from causing unacceptable releases or radionuclides.
retardation	A reduction in the velocity of radionuclide movement through the environment due to reversible adsorption on an immobile matrix. Soils often retard movement of waterborne nuclides, and the degree of retardation can be quantified.
retrievability	The capability to remove waste from where it has been stored.

risk	With respect to radiation protection, the probability that a given individual will incur any given deleterious stochastic effect as a result of radiation exposure.
rock	To the geologist, any mass of mineral matter, whether consolidated or not, which forms part of the Earth's crust. Rocks may consist of only one mineral species, in which case they are called monomineralic, but they more often consist of an aggregate of mineral species.
routine discharges	Planned and controlled release of radionuclides to the environment. Such releases will meet all restrictions imposed by appropriate regulatory authorities.
safety	Protection of all persons from undue radiological hazard.
safety analysis	The analysis and calculation of the hazards (risks) associated with the implementation of a proposed activity.
safety assessment	A comparison of the results of safety analyses with acceptability criteria, its evaluation, and the resultant judgments made on the acceptability of the system assessed.
salt	A geological formation containing mainly halite (NaCl) with smaller inclusions of other minerals, usually the chloride or sulfate derivatives of the alkaki or alkaline earth elements. Salt formations occur as bedded or domal deposits. These dry, stable formations are considered to be good hosts for deep underground waste repositories.
salt dome	A dome-like salt structure resulting from the upward movement of a salt mine.
sea-bed disposal	Placement of waste packages in deep ocean sediments.
sea dumping (disposal)	The practice of periodically dumping shiploads of drummed, solidified waste at specified locations in the ocean.

separations (partitioning)	Separations in this report implies separation of a process stream into two or more components for waste management purposes; rather complete separations are usually envisaged.
shallow ground disposal	Disposal of radioactive waste, with or without engineered barriers, above or below the ground surface, where the final protective covering is a few meters thick. Some member nations of the International Atomic Energy Agency consider shallow ground disposal to be a mode of storage rather than a mode of disposal.
shipping cask (transport cask)	A heavy protective container that shields and contains radioactive materials, dissipates heat, and prevents criticality during transport and handling.
short-lived nuclide	For waste management purposes, a radioactive isotope with a half-life shorter than about 30 years (e.g., ^{137}Cs, ^{90}Sr, ^{85}Kr, ^{3}H).
short-lived waste	Waste that will decay to a level that is considered to be insignificant from a radiological viewpoint, in a time period during which administrative controls can be expected to last. Such waste can be determined by radiological assessment of the storage or disposal system chosen (see *long-lived waste*).
shutdown	Actions taken at a repository after disposal operations have ceased, in order to prepare the facility for abandonment. This includes decommissioning of ancillary facilities and sealing the repository. Shutdown may occur immediately or after a period of surveillance following the final emplacement of waste.
sievert (Sv)	The unit of dose equivalent (1 Sv = 100 rem).
site	The area containing a nuclear installation (e.g., waste repository) that is defined by a boundary and that is under effective control of the implementing organization.

site confirmation	Actions involved in establishing the suitability of an intended repository site. Extensive onsite investigations will be made to assure its capability for achieving all performance criteria, especially radiation protection requirements. The type of waste to be emplaced and the proposed repository design are of importance in this assessment.
siting	The process of selecting a suitable site for an installation, including appropriate assessment and definition of the related design bases.
solidified waste, radioactive	Liquid waste or otherwise mobile waste materials (ion exchange resins, etc.) that have been immobilized by incorporation (either physical or chemical) into a solid matrix by some specific treatment.
solid waste, radioactive	Untreated waste that possesses physical properties commonly associated with the solid state. Animal carcasses are usually considered to be in this category.
sorption	A broad term referring to reactions taking place within pores or on the surfaces of a solid. Its use avoids the problem of technical distinction between absorption and adsorption reactions. *Absorption* is generally used to refer to reactions taking place largely within the pores of solids, in which case the capacity of the solid to absorb is proportional to its volume. *Adsorption* refers to reactions taking place on solid surfaces, so that the capacity of a solid is proportional to its effective surface area. An example of the latter process is *ion exchange*, whereby ions occupying charged sites on the surface of the solid are displaced by ions from solution.
speciation	A term that refers to the chemical form(s) and properties of a radionuclide under a particular set of environmental conditions (pH, eH, ligands present, etc.). Speciation study is valuable because the environmental behavior of a nuclide is largely determined by its chemical form.
spent fuel	Irradiated fuel units not intended for further reactor service.
subsequent control	Any long-term safety measures, such as land-use restrictions, imposed to assist in achieving repository safety after shut-down and sealing has been completed and after the operating license for the repository has been cancelled.

subsidence	Sinking or caving in of the ground surface. This results from the inability of the upper layers of the Earth's crust to support their own mass, or that mass with additional surface load, over an area containing poorly compacted material or voids. Such voids can be man-made, as in the case of mines.
surface water	Water that fails to penetrate into the subsoil and flows along the surface of the ground, eventually entering a lake, a river, or the sea.
surveillance	All planned activities performed to ensure that conditions at a nuclear installation remain within the prescribed limits. For a waste repository, surveillance continues well past the periods of operation and closure.
synroc	The name given to a group of specially formulated zirconium-based ceramics that were originally developed by Australian scientists for immobilizing high-level waste.
tailored ceramic	A ceramic material whose composition has been specially formulated to optimize the incorporation of a particular set of radionuclides into the crystalline matrix. The composition of the waste form is thus tailored to fit a particular waste stream (See *ceramic materials; synroc*).
thermal gradient	A quantitatively measurable change in sensible heat as a function of distance. In a deep geological repository, the thermal gradient produced by radioactive decay heat can potentially alter the host rock and water-flow paths.
thermal loading	The quality of heat-generating materials placed in a given area or volume; units are power per area or per volume, respectively.
topography	(1) The configuration of (a portion of) the Earth's surface, including its relief and relative positions of its natural and man-made features. (2) The practice of graphical representation of the same.
transmissivity, hydraulic	Rate at which water is transmitted through a unit width of aquifer under a unit hydraulic gradient. It is expressed as the product of the hydraulic conductivity and the thickness of the saturated portion of the aquifer.

transmutation	Transmutation in this report generally implies the conversion of long-lived radioactive species either to short-lived or to stable species by means of nuclear processes.
transuranic (TRU) waste	Waste containing quantities of nuclides having atomic numbers above 92 in excess of agreed limits. The limits are established by national regulatory bodies. (See *alpha-bearing waste*.)
treatment of waste	Operations intended to benefit safety or economy by changing the characteristics of the waste. Three basic treatment concepts are —volume reduction, —removal of radionuclides from the waste, and —change of composition (see *conditioning of waste*).
tuff	One of a series of pyroclastic rocks composed of consolidated ash from fragmental volcanic material blown into the atmosphere by volcanic activity.
underground disposal	Disposal of waste at an appropriate depth below the ground surface.
uptake	Amount of radioactive material absorbed into the extra cellular fluids. It is also used to denote the process.
validation	A process carried out by comparison of model predictions with independent field observations and experimental measurements. A model cannot be considered validated until sufficient testing has been performed to ensure an acceptable level of predictive accuracy. (The acceptable level of accuracy is judgmental and will vary depending on the specific problem or question to be addressed by the model.)
vault	An above-or below-ground reinforced concrete structure that contains an array of storage cavities, each of which could hold one or more spent fuel units or waste packages. Shielding is provided by the exterior of the structure. Heat removal is principally by forced or natural movement of gases over the exterior of the cavities. Heat rejection to the atmosphere is either direct or via a secondary cooling system.

verification	A mathematical model, or the corresponding computer code, is verified when it is shown that the code behaves as intended, that is, that it is a proper mathematical representation of the conceptual model and that the equations are correctly encoded and solved.
vermiculite	A group of micaceous clay minerals closely related to chlorite and montmorillonite and having the general formula $(Mg,Fe,Al)_3(Al,Si)_4 0_{10}(OH)_2\cdot 4H_2 0$. Because of its adsorptive properties, vermiculite is often used in packaging small quantities of liquid waste.
vitrification	Any process of converting materials into a glass or glass-like form.
volume reduction	A treatment that decreases the physical volume of a waste. Volume reduction is used to facilitate subsequent handling, storage, transportation, or disposal of the waste. Typical treatments are mechanical compaction, incineration, or evaporation. Volume reduction results in a corresponding increase in radionuclide concentration.
volume reduction factor	The ratio of the volumes of radioactive waste prior to and following treatment. In concentration processes, the factor is greater than one; in dilution systems, the factor is less than one.
waste arisings	Radioactive wastes generated by any stage in the nuclear fuel cycle and by any other activity dealing with radioactive materials.
waste disposal	See *disposal*.
waste form	The physical and chemical form of the waste materials (e.g., liquid, in concrete, in glass) without any packaging.
waste glass	The vitreous product that results from incorporating waste into a glass matrix.
waste package	The waste form and any container(s) as prepared for handling, transport, treatment, conditioning, storage, and disposal of waste.

Appendix O

Acronyms And Abbreviations

AB	Actinide-Burning
AEA	Atomic Energy Act (1984)
AEC	Atomic Energy Commission
ALMR	advanced liquid-metal reactor
ALWR	advanced light-water reactor
ANL	Argonne National Laboratory
ATW	accelerator transmutation of waste
BNL	Brookhaven National Laboratory
BWR	boiling water reactor
CAA	Clean Air Act
CCDF	complementary cumulative distribution function
CCL	coupled cavity linac
CEA	Council for Energy Awareness (U.S.)
CEC	Commission of the European Communities
CERCLA	Comprehensive Environmental Response, Compensation, and Liability Act
CF	capacity factor
CMPO	carbonmoyl methyl phosphine oxide
CSFR	Czech and Slovak Federal Republic
CTH	Chalmers University of Technology
CURE	Clean Use of Reactor Energy
DF	decontamination factor
DHLW	Defense High-Level Waste
DMDBTDMA	dimethyl dibutyl tetradecyl methyl amide
DOE	Department of Energy
DOE/NE	Department of Energy/Nuclear Energy Office
DS	double-shell (adj.)
DTL	drift tube linac
DWPF	Defense Waste Processing Facility (Savannah)
ECU	European currency units
EIA	Energy Information Administration
EPA	Environmental Protection Agency
EPRI	Electric Power Research Institute
ERA	Energy Reorganization Act
ERDA	Energy Research and Development Administration
FEIS	Final Environmental Impact Statement
FFTF	Fast Flux Test Facility

FP	Fission Product
FWPAC	Federal Water Pollution Control Act
GCDF	Greater Confinement Disposal Facility
GE	General Electric
GESMO	Generic Environmental Impact Statement on Mixed Oxide
GTCC	Greater-Than-Class-C
HEU	high enriched uranium
HFIR	high flux isotope reactor
HLW	high-level waste
HM	heavy metal
HSWA	Hazardous and Solid Waste Amendments of 1984
HTGR	high-temperature gas-cooled reactor
HWVP	Hanford waste vitrification plant
HWR	heavy-water reactor
IAEA	International Atomic Energy Agency (U.N.)
ICRP	International Commission on Radiation Protection
IFR	integral fast reactor
ILW	intermediate-level waste
INEL	Idaho National Engineering Laboratory
INFCE	International Fuel Cycle Evaluation
LAMPF	Los Alamos Meson Physics Facility
LAMPRE	Los Alamos Molten Plutonium Reactor Experiment
LANL	Los Alamos National Laboratory
LEU	low enriched uranium
LGR	liquid graphite reactor
LLW	low-level wastes
LMFBR	liquid-metal fast breeder reactor
LMFR	liquid-metal fast reactors
LMR	liquid-metal reactor
LWR	light-water reactor
MA	minor actinide
MGD	Mined Geologic Disposal
MOX	mixed oxide
MRS	monitored retrieval storage
MSRE	Molten Salt Reactor Experiment
MTHM	metric tons of heavy metal
MTIHM	metric tons of initial heavy metal
MTU	metric tons uranium
MUF	material unaccounted for
MWD/MT	megawatt-days per metric tons
NAS	National Academy of Sciences
NCRP	National Council on Radiation Protection and Measurements
NEA	Nuclear Energy Agency

NEPA	National Environmental Policy Act
NPDES	National Pollutant Discharge Elimination System
NPT	Nuclear Nonproliferation Treaty (1978)
NRC	Nuclear Regulatory Commission
NWPA	Nuclear Waste Policy Act (1982)
NWPAA	Nuclear Waste Policy Amendments Act (1987)
OECD	Organization of Economic Cooperation and Development
OMEGA	Option Making Extra Gains from Actinides and Fission Products (Japan)
ORNL	Oak Ridge National Laboratory
P-T	Partitioning and Transmutation
PBR	particle bed reactor
PNL	Pacific Northwest Laboratory
PRA	probabilistic risk assessment
PRISM	power reactor, innovative, small module
PSI	The Paul Scherrer Institute
PUREX	plutonium and uranium recovery by extraction
PWR	pressurized water reactor
RCRA	Resource Conservation and Recovery Act
REDOX	oxidation-reduction chemistry process
RF	radio frequency
RFQ	radio frequency quadrop
S&T	separations and transmutation
SAF	secure automated fabrication
SAFR	sodium advanced fast reactor
SAR	Safety Analysis Report
SARA	Superfund Amendments and Reauthorization Act of 1986
SCP	Site Characterization Plan
SRS	Savannah River Site
SS	single-shell
STATS	Committee on Separations and Transmutation Systems
SWU	separative work unit
TBP	tributyl phosphate
THORP	Thermal Oxide Reprocessing Plant (U.K.)
TRU	transuranic (wastes/elements)
TRUEX	transuranic extraction
WHC	Westinghouse Hanford Company
WIPP	Waste Isolation Pilot Plant

Appendix P

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Index