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COMMITTEE ON NUCLEAR SCIENCE

The Radiochemistry of Carbon, Nitrogen, and Oxygen

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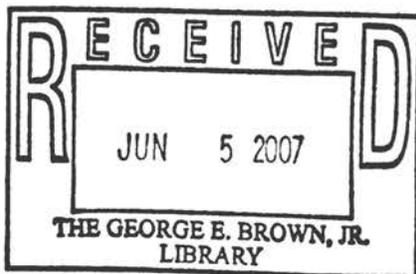
Subcommittee on Radiochemistry

National Academy of Sciences—National Research Council

FOREWORD

The Subcommittee on Radiochemistry is one of a number of subcommittees working under the Committee on Nuclear Science within the National Academy of Sciences - National Research Council. Its members represent government, industrial, and university laboratories in the areas of nuclear chemistry and analytical chemistry

The Subcommittee has concerned itself with those areas of nuclear science which involve the chemist, such as the collection and distribution of radiochemical procedures, the establishment of specifications for radiochemically pure reagents, availability of cyclotron time for service irradiations, the place of radiochemistry in the undergraduate college program, etc.

This series of monographs has grown out of the need for up-to-date compilations of radiochemical information and procedures. The Subcommittee has endeavored to present a series which will be of maximum use to the working scientist and which contains the latest available information. Each monograph collects in one volume the pertinent information required for radiochemical work with an individual element or a group of closely related elements.

An expert in the radiochemistry of the particular element has written the monograph, following a standard format developed by the Subcommittee. The Atomic Energy Commission has sponsored the printing of the series.

The Subcommittee is confident these publications will be useful not only to the radiochemist but also to the research worker in other fields such as physics, biochemistry or medicine who wishes to use radiochemical techniques to solve a specific problem.

W. Wayne Meinke, Chairman
Subcommittee on Radiochemistry

iii

INTRODUCTION

This volume which deals with the radiochemistry of carbon, nitrogen and oxygen is one of a series of monographs on radio-chemistry of the elements. There is included a review of the nuclear and chemical features of particular interest to the radiochemist, a discussion of problems of dissolution of a sample and counting techniques, and finally, a collection of radio-chemical procedures for the element as found in the literature.

The series of monographs will cover all elements for which radiochemical procedures are pertinent. Plans include revision of the monograph periodically as new techniques and procedures warrant. The reader is therefore encouraged to call to the attention of the author any published or unpublished material on the radiochemistry of carbon, nitrogen and oxygen which might be included in a revised version of the monograph.

The Radiochemistry of Carbon,
Nitrogen, and Oxygen

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I. INTEREST AND USES OF CARBON, NITROGEN, AND OXYGEN TO THE RADIOCHEMIST

This review of carbon, nitrogen, and oxygen chemistry of interest to the radiochemist is limited in scope for a number of reasons. No work on C^{14} will be reviewed since that nuclide, its uses and applications, seem to be almost completely within the province of the organic chemist. The remaining useful radioactive isotopes of the elements in question, 20.4-min. C^{11}, 10.0-min. N^{13}, and 2-min. O^{15}, have half-lives so short that their use as tracers has been severely restricted by time limitations. In addition, since these three nuclides are neutron-deficient, charged-particle accelerators are necessary to produce reasonable amounts, further limiting the usefulness of these nuclides. However, as will be described in Section IV a method exists whereby at least N^{13} may be produced in reasonable quantities in neutron reactors.

Finally, the large number of possible oxidation states and resulting complex chemistry of carbon and nitrogen allow many interesting experiments in the field of hot-atom chemistry.[1,2] Discussion of these problems will also be avoided except where pertinent information is necessary to ensure

1

isotopic mixing of carrier and tracer atoms during the course of a chemical separation process.

C^{11}

(3/2⁻) ——— C^{11} ——— (t = 20.4 min)

β^+ (99+ %)

EC (0.19%)

E_{β^+} = 0.97 Mev

(3/2⁻) ———↓———

B^{11}

N^{13}

(1/2⁻) ——— N^{13} ——— (t = 10.0 min)

β^+ (100%)

E_{β^+} = 1.19 Mev

(1/2⁻) ———↓———

C^{13}

O^{15}

(1/2⁻) ——— O^{15} ——— (t = 124 sec)

β^+ (100%)

E_{β^+} = 1.72 Mev

(1/2⁻) ———↓———

N^{15}
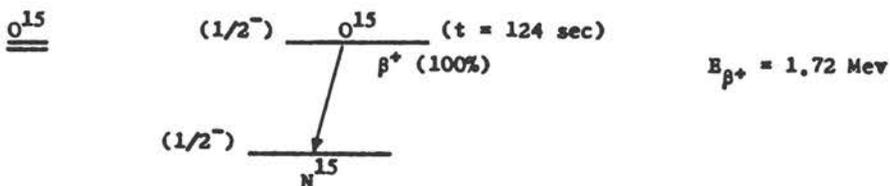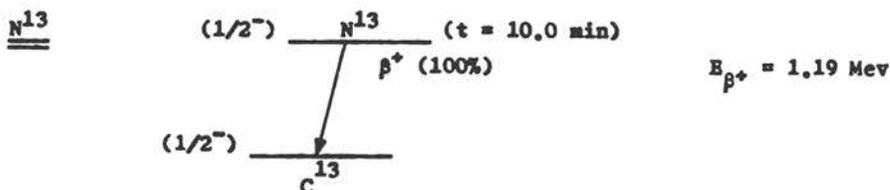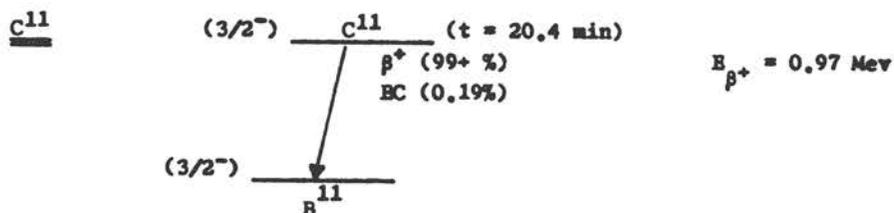
Fig. 1. Nuclear properties of C^{11}, N^{13}, and O^{15}[3]

The main interest in C^{11}, N^{13}, and O^{15} thus far to the nuclear chemists has been in the determination of the yields of these nuclides in nuclear reactions resulting from the irradiation of complex target nuclei with charged particles. As long as the target material is an element no heavier than aluminum the yields of these nuclides may be determined by gross

activity measurement of the irradiated sample and resolution of the resulting complex decay curve into its components. Indeed most of the references to C^{11} and N^{13} refer to this type of experiment. However, when one desires information about the yields of C^{11} and N^{13} from heavier targets such as copper and silver the gross decay curve is far too complex to resolve into its various components and the elements desired must be chemically separated from the target and interfering activities.

With the advent of high-energy accelerators there is great interest in yields of relatively light products such as C^{11} and N^{13} and thus the radiochemistry of these elements is of renewed interest. In addition the ever increasing number of laboratories having access to charged-particle accelerators will undoubtedly bring about new investigations making use of these short-lived tracers. It is interesting to note that whereas Ruben and co-workers[4-6] made excellent use of C^{11} in studying the mechanism of reactions $\sim$ 20 years ago, little has been done with this isotope in the field of reaction kinetics since then.

II. REVIEW OF THOSE FEATURES OF CARBON, NITROGEN, AND OXYGEN CHEMISTRY OF INTEREST TO THE RADIOCHEMIST

Carbon.—The main problems regarding the chemistry of carbon which the radiochemist faces are those of isotopic mixing of carrier and tracer atoms, complete conversion to a single known oxidation state, and purification and separation of the resulting compound from the reaction mixture and interfering activities. The problem of the knowledge of the oxidation state or states of the radioactive carbon atoms formed during an irradiation and present after dissolution of the target is a complex one. The usual procedure is to dissolve the target in the presence of one or more lower oxidation states of carbon and oxidize the carbon compounds to CO_2. Thus it is hoped that all active carbon atoms will undergo mixing with the carrier atoms during some phase of the initial chemical treatment. The CO_2 is then freed of possible volatile radioactive contaminants such as the rare gases, converted to a

3

measurable form, and assayed. The chief difficulty is that one is seldom sure that all of the active carbon atoms have followed the chemistry of the carrier.

To oxidize all carbon compounds to CO_2 there seem to be two lines of attack. One is to dissolve the target material in a very strongly oxidizing medium and assume that all the carbon comes off as CO_2. The other is to sweep the volatile carbon compounds through a copper or nickel oxide combustion furnace in an effort to bring about the oxidation to CO_2. Sharp[7] found that to obtain complete and reproducible yields both methods had to be combined and in fact a very elaborate system was evolved.

Nitrogen.—As with carbon the difficulties in handling nitrogen arise from the large number of oxidation states in which N^{13} may find itself after irradiation and dissolution of the target. Reference 8 is an excellent source of information about the chemical behavior of nitrogen. In addition, with a 10-min. compared to a 20-min. half-life, the time available for adequate purification is even shorter for nitrogen than for carbon. However, as will be described in detail Dostrovsky and Hudis[9] have found, at least in the high-energy proton irradiation of aluminum, zinc, indium, lead, and uranium foils, that the N^{13} atoms are all present in the NH_3 fraction if the metal foils are dissolved in acid under reducing conditions. Those foils which would dissolve in strong NaOH — aluminum and zinc — were also found to yield all the N^{13} as NH_3. The chemical separation procedure then reduces to NH_3 purification from volatile activities such as carbon compounds, rare gases, and possibly AsH_3, SbH_3 etc. Detailed descriptions of the various dissolution processes are given. It would be interesting to see if under identical conditions of target solution one could obtain all C^{11} activity as methane.

Oxygen.—As far as the author can tell no one has been brave or foolhardy enough to attempt to separate chemically and purify 2-min. O^{15} from any target heavier than aluminum. From aluminum the separation reduces to

4

the elimination of N^{13} and Ne^{23} activities, all other nuclides having half-lives short or long enough not to interfere with the determination of O^{15}. From a water target swept with oxygen gas, Katcoff and Hudis[10] found that a liquid O_2 trap and two CaF_2 drying tubes followed by trapping of the oxygen in a liquid nitrogen cooled trap was sufficient to yield pure 2-min. O^{15} activity.

III. MEASUREMENT TECHNIQUES

The radioactive decay properties of C^{11}, N^{13}, and O^{15} are quite similar; each emits a single energetic positron with no accompanying nuclear gamma ray.

Beta Measurement.—Direct measurement of the positron activity is possible and has been used by a number of workers. The single positron branch and high energy make it relatively simple to calibrate the Geiger or proportional counter set-up for the specific nuclide in question when absolute disintegration rates are desired. For the determination of C^{11} activity the usual procedure is to bubble the active CO_2 through a $Ba(NO_3)_2$ solution and quickly filter, dry, and count the $BaCO_3$ precipitate. Likewise, NH_3 can be precipitated as $(NH_4)_2PtCl_6$ or as $NHg_2I \cdot H_2O$. Chackett et al.[11] recovered NH_3 in a measurable form very quickly by passing the gas through a filter paper moistened with dilute sulfuric acid.

At least one worker has assayed C^{11} as $C^{11}O_2$ incorporated into the counting gas mixture of a beta-proportional counter. This is one of the more popular methods of counting C^{14} activity and details of the sample preparation and counting procedures may be found in Reference 12. The chief advantage of this technique is, of course, that one attains excellent counting efficiencies — almost 100%.

The one great drawback of both of these procedures when one considers the short half-lives involved is the time required at the end of the separation to prepare either solid or gaseous samples suitable for beta counting.

Gamma Measurement.—The detection of the annihilation radiation by means of scintillation counters is a technique which promises to be by far the easiest and fastest method for the measurement of these isotopes. With the large NaI crystals presently available and the presence of two gammas per disintegration very high counting efficiencies are obtainable. In addition the very slight dependence of counting efficiency on sample thickness or physical state enables one to measure the sample immediately following the last chemical purification step.

For example, in a search for O^{20}, Katcoff and Hudis[10] made use of the O^{15} present to check on the oxygen chemistry. The final step in the purification was the trapping of oxygen in a liquid nitrogen trap. A gamma counter was placed on top of the trap and counting commenced about thirty seconds after the end of the irradiation.

Similarly, Dostrovsky and Hudis[9] have measured the gamma radiation from N^{13} in various ways, (a) solutions of boric acid in which NH_3 had been neutralized, (b) traps of Li metal which were used as getters for the recovery of N_2 and N_2O, and (c) activated charcoal traps which had been cooled to liquid nitrogen temperatures and used to trap N_2 and the rare gases.

Thus it can be seen that almost any part of the recovery and purification system which can be removed and placed on or in a scintillator makes a convenient counting sample. The particular counting set-up can be calibrated at some later time using a standard which emits about 510 kev gamma radiation and has a more convenient half-life.

IV. METHODS OF PRODUCTION OF C^{11}, N^{13}, and O^{15}

These three nuclides are all neutron-deficient and can be conveniently produced only by high-energy neutron or charged particle reactions. Table I gives a list of the most useful reactions available. It may be seen from the table that these nuclides may be made with quite low-energy charged particles. However, the fact that one may quickly label a given compound and eliminate precious time in separating the desired activity from the target material

6

Table I. Nuclear Reactions Useful for the Production of C^{11}, N^{13}, and O^{15}

	Target	Relative Isotopic Abundance %	Particle	Reaction	Q (Mev)[13]
C^{11}	Be^9	100	α	$\alpha, 2n$	-24.4
	B^{10}	18.8	d	d, n	+ 6.5
	B^{11}	81.2	p	p, n	- 2.8
	B^{11}	81.2	d	d, 2n	+ 5.3
	C^{12}	99	n	n, 2n	-18.7
N^{13}	B^{10}	18.8	α	α, n	+ 1.1
	B^{11}	81.2	α	$\alpha, 2n$	-10.3
	C^{12}	99	d	d, n	- 0.3
	N^{14}	$\sim$ 100	n	n, 2n	-10.5
O^{15}	C^{12}	99	α	α, n	- 8.5
	N^{14}	$\sim$ 100	d	d, n	+ 5.1
	O^{16}	$\sim$ 100	n	n, 2n	-15.7

favors the production of these nuclides by fast neutron irradiation of the parent compound whenever possible unless too much target decomposition occurs due to irradiation damage.

For example, if one irradiates NH_4NO_3 with fast neutrons, both nitrogen atoms are tagged and the chemistry of NH_4^+ and NO_3^- ions may be followed. Addition of Br_2 to the target solution yields N_2 tagged with N^{13} allowing one to investigate the chemistry of N_2.

N^{13} is the only nuclide under discussion which can be produced by neutrons available from low-energy, charged particle accelerators such as a Cockroft-Walton or a Van de Graaff. The $H^3(d, n)He^4$ reaction yields $\sim$14 Mev neutrons, sufficiently energetic to produce the (n, 2n) reaction on N^{14}. Unfortunately quite high energy neutrons are required to produce

C^{11} and O^{15} by the n,2n reaction and these very fast neutrons can be created in quantity only with high intensity, high energy particle accelerators such as cyclotrons.

With the increasing availability of neutron reactors another scheme capable of producing fast neutrons might be mentioned here. Weiss[14] has shown that by irradiation of Li^6D with thermal neutrons, 14 Mev neutrons can be obtained in the ratio of one 14 Mev neutron to every 10^6 thermal neutrons. The nuclear reactions are (a) $Li^6(n,H^3)He^4$ followed by (b) $H^2(H^3,n)He^4$. That is to say the tritons produced in reaction (a) collide with the deuterons of the LiD molecules to produce 14 Mev neutrons by reaction (b). In this way it is possible to obtain usable quantities of N^{13} activity with a nuclear reactor.

V. COLLECTION OF DETAILED RADIOCHEMICAL PROCEDURES

It should be pointed out that the following procedures have been quoted verbatim from the original references and except for Procedure #1 of nitrogen none have been checked by the author. It seems obvious, in carbon Procedures 3, 4, and 5 that very few if any checks have been made to determine whether or not all of the C^{11} produced had been converted to an insoluble carbonate precipitate. From the precautions taken by Sharp[7] to obtain reproducible C^{11} yields we would hesitate to place too great faith in the simpler carbon procedures, at least from targets heavier than aluminum.

CARBON

Procedure 1. Yield of C^{11} from High-Energy Proton Irradiation of Al.

Source: K. Chackett et al., Proc. Phys. Soc. (London) 69A, 43 (1956).

This activity appears in the gas phase on the dissolution of the aluminum target foil in boiling sodium hydroxide solution. It is swept through a CuO combustion train, the CO_2 being trapped in caustic soda and counted as $BaCO_3$.

Procedure 2. Yield of C^{11} from High-Energy Proton Irradiation of Al.
Source: K. Chackett et al., Phil. Mag. 46, 1 (1955).

The bombarded aluminum foils were dissolved in sodium hydroxide solution
in a small reaction vessel through which passed a slow stream of air saturated
at room temperature with ethanol vapor. The gas stream was roughly dried by
bubbling through concentrated sulfuric acid and was then passed over CuO at
600°C. In this way the C^{11} originally present in the foils was converted in
the gas stream to $C^{11}O_2$ in the presence of a suitable quantity of CO_2 derived
from the ethanol. The gas was then bubbled through two centrifuge cones
containing sodium hydroxide solution, which absorbed all the CO_2. These
cones were detached from the apparatus and $BaCO_3$ precipitated by the addition
of $Ba(OH)_2$. The $BaCO_3$ was washed, mounted, and counted under an end-window
G-M counter in the usual way. It carried the C^{11} activity practically
quantitatively and completely free from all other activities.

Procedure 3. Yield of C^{11} from the High-Energy Proton Irradiation of Ag.
Source: B. V. Kurchatov et al., Conf. Acad. Sci. U.S.S.R., Peaceful Uses
       Atomic Energy, Moscow (1955). U.S. Atomic Energy Commission Transl.,
       2435, Part 2, 111-24 (1955).

The silver foil was melted in a hydrogen atmosphere before irradiation.
After irradiation the foil was dissolved in nitric acid, gases bubbled over
hot CuO into 20% NaOH. Boiling was continued for ten minutes with passage of
air. The C^{11} was counted as $BaCO_3$.

Procedure 4. Yield of C^{11} from the High-Energy Proton Irradiation of Co.
Source: B. Belmost, U.S. Atomic Energy Commission Document N.Y.O. 3198.

Five mg of carbon as $NaHCO_3$ was added dropwise to the reaction vessel
and the evolved gas was led into a freshly prepared, carbonate-free solution
of NaOH. The rate of addition of the carrier was adjusted so that about half
was added after the target was dissolved. Nitrogen was bubbled through the
system to prevent the base from being sucked back into the reaction vessel
during pressure fluctuations. The sodium hydroxide solution was diluted to

100 ml and freshly prepared barium nitrate was added. The barium carbonate precipitate was washed with water and mounted.

Procedure 5. Yield of C^{11} from the High-Energy Proton Irradiation of Ag.

Source: P. Kofstad, UCRL 2265.

The silver target was dissolved in a distillation flask. Any escaping gases were collected in a 1 M solution of NaOH saturated with $Ba(OH)_2$ in an ice bath. The nitric acid solution was made slightly basic with KOH and carbon carrier as Na_2CO_3 was added. A solution of hypochlorite was added and the solution boiled for a few minutes to oxidize all the active carbon to the carbonate form. Any escaping gases were continually caught in the saturated $Ba(OH)_2$ solution. After oxidation of the carbon the solution was made acidic with HCl. Carbon dioxide-free air was bubbled through for a few minutes while the solution was gently warmed to drive out all the CO_2 gas. $BaCO_3$ precipitated. This precipitate was transferred to a distillation flask, silver and bromide carriers were added, phosphoric acid and a few ml of saturated boric acid were added. The solution was gently warmed while CO_2-free air was passed through, catching the CO_2 in a saturated solution of $Ba(OH)_2$ in an ice bath. The final $BaCO_3$ precipitate was finally washed with a dilute solution of NH_4OH, water, and acetone, dried at $110^{\circ}C$ and weighed.

Procedure 6. Yield of C^{11} from the High-Energy Proton Irradiation of Al, Cu, In, Bi, and U.

Source: R. Sharp (private communication).

Night before run:

1. Fill carrier gas cell CG with 5 cc-atm of 2:1 CO_2:CH_4 (containing 2% Xe for targets above Te).

*2. Fill bubbler BT with 50 cc saturated $Ba(OH)_2$ solution.

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*Denotes steps done only for targets above Te.

3. Fill scrubbing tube ST half full at 80% H_2SO_4 and attach target solution flask TF+CC to vacline with V5 open. Attach condenser hoses.

4. With counting tube CT on line, pump out whole system overnight with opened V5, 6, 7, 8, 10↓, 11←, 12↓, 13, N, 14↓, 15, 17←, 18, 19, 20, 21, 22, 24, 25↑, 26 and closed V1, 2, 3, 4, 9, 16, 23.

Morning before run:

5. Close V7, 13, 17, pump out counting tube to $< 10^{-5}$ mm.

6. Close V14↑, N, 17, 19, 22, 24, 25↓.

7. Set NiO_2 furnace NF to 950°C, PbO_2/CuO trap PC to 200°C.

8. Immerse trap I in dry ice-acetone.

9. Open V9 briefly to 1 atm on gauge GA.

10. Replace carrier gas cell CG with shorting line connected between joints A and B. Open V1, 13 and adjust N to 50 cc/m on flow meter FM.

*11. Open V10↑, 12↑, flush bubbler BT 3 min. Close V10↓, 12↓ to bypass bubbler.

12. Make fresh phosphoric acid grease. Turn on water in condenser CC.

Run:

13. Close V1, 6, open V9. Remove target solution flask TF+CC. Attach carrier gas cell CG to target solution flask.

14. Place weighed target in cup TF, wipe V4 and joint C and apply fresh phosphoric acid grease. Attach cup TF to target solution flask. Attach target solution flask to vacline. Attach heat shield to Joint C.

15. Open V1, 6. Close V8.

16. Close V21, 13, 9. Open V7. Pump to 1 mm pressure. For targets below Te, immerse trap II in liquid N_2.

17. Close V7, 26. Open V21.

18. Open V10↑, 12↑.

19. Dissolve Target:

(a) Open V2, 3. Fill target solution flask with carrier gas and oxygen to about 50 cm pressure.

(b) Close V2, 5. Dissolve the target and add combustion fluid.

20. Boil the combustion fluid containing the dissolved target one min.

21. Open V13, 26, 8, 2.

For targets below Te:

A22. Sweep system 5 min at 50 cc/min on flow meter FM.

A23. Sweep 5 min at 100 cc/min.

A24. Close V2, pump system to 19 cm pressure while adjusting N to maintain sweep rate of $\sim$ 50 cc/min rate on FM.

A25. Double sweep rate to 100 cc/min and pump to 3 cm pressure.

For targets above Te:

B22. Sweep system 3 min at 75 cc/min. Close V2, pump out system to 12 cm pressure while adjusting N to maintain $\sim$ 75 cc/min rate on FM. Close N. Open V2 slowly, filling system with O_2 to one atm. Open N to 75 cc/min rate on FM. Close V2.

B23. Repeat B22 two more times.

B24. Immerse trap II in liquid N_2. Reduce sweep rate to $\sim$10 cc/min with N. Add 20 cc of 20% H_3PO_4 to bubbler BT through filling funnel BF.

B25. Open V2, sweep 3 min at 75 cc/min, close V2, pump to 12 cm pressure maintaining 75 cc/min sweep rate on FM.

---

26. Close V13, 14$\downarrow$, 15, 20, 21. Open N, 22, 24, 25$\uparrow$.

27. Pump to 10^{-4} mm pressure opening V18, 19 when pressure reaches 10^{-3} mm.

28. Turn off Philips gauge. Close V22. Transfer $C^{11}O_2$ to counter by thawing trap II with hair dryer while immersing bottom end of counting tube in liquid N_2. Close V19.

29. Thaw $C^{11}O_2$ in counting tube with hair dryer.

30. Open V16. Open V19 briefly to fill counter to one atm with P10. Close V19.

31. Close V16, 18.

32. Start counting 6 min after filling.

Remarks

4. V denotes stopcock. V14↓ denotes three way cock turned to direct flow into lower leg in schematic of vacline, V14↑ upper leg, etc.

8. At least one quarter of traps I and II must be above level of refrigerant for efficient operation.

10. Flow meter FM operates at 1 cm pressure but is calibrated to read STP cc/m flow rate in system before expansion valve N.

12. Cover HPO_3 pellets with 85% H_3PO_4 and boil to complete solution. Cool.

14. The heat shield for joint C is a flat plate of 1/16" aluminum which slips over TF and is held in place by springs. This keeps the grease on joint C from melting when a Bunsen burner is applied to TF.
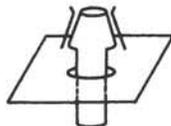
15. After step 15, the stopcocks should be set as follows: Opened V1, 5, 6, 9, 11←, 13, N, 14↑, 15, 18, 20, 21, 25↓, 26. Closed V2, 3, 4, 8, 10↓, 12↓, 16, 17, 19, 22, 23, 24.

16. Pumping to 1 mm removes the air introduced when the target solution flask was reattached.

19. (a) Target solution flask is filled with oxygen to a pressure such that the total pressure in the flask after dissolving the target is about 70 cm. More would cause the joints to open on boiling the combustion fluid; less would allow the fluid to boil at a lower temperature thus reducing the speed of oxidation.

(b) The targets are dissolved as follows:

Aluminum: 7-1/2 cc A added to cup TF through funnel FF.

Heat to solution of Al. Add 7-1/2 cc B, 1/4 gm KIO_3.

Copper: 1 cc conc HNO_3 added to TF, dissolving the Cu.

10 cc plus 1/4 gm KIO_3 added.

Indium: Same as copper.

Bismuth: 1-1/2 cc 75% HNO_3 added to TF, dissolving the Bi,

10 cc C plus 1/4 gm KIO_3 added.

Uranium: 1 cc conc HNO_3 added to TF, warm gently to dissolve

the U. 10 cc C + 1/4 gm KIO_3 added.

A is 33 cc 85% H_3PO_4 plus 22 cc conc (36M) H_2SO_4 plus 45 cc fuming H_2SO_4 (30% SO_3) to make 100 cc total.

B is A + 10.4 g CrO_3.

C is A + 5.2 g CrO_3.

A22, B22. Sweeping by pumping down several times to a fraction of an atmosphere is a faster method than sweeping continuously at one atmosphere pressure due to dead volumes in the apparatus. The best sweeping procedure will depend on the configuration of the apparatus and should be checked for efficiency by manometric or radioassay. The extensive sweeping used for targets above Te is necessary due to the high solubility of Xe and Rn in water and the consequent difficulty in purging them from the bubbler solution. For cosmotron bombardments, the sweeping used in the recipe gave a 1-3% rare gas activity tail in the C^{11} decay curves. Higher decontamination may be necessary for low energy bombardments. There is room for improvement in the Rn + Xe scavenging method. A much more efficient and probably faster separation of $C^{11}O_2$ from Rn + Xe could probably be had via gas chromatography using one of the excellent commercial devices now available.

B24. Indicator in the $Ba(OH)_2$ solution can be used to insure neutralization by the H_3PO_4. $Ba_3(PO_4)_2$ precipitates on first addition of phosphoric acid but quickly dissolves in excess of the reagent.

30. P-10 pressure in line before V16 is regulated to 76 cm with a reducing valve.
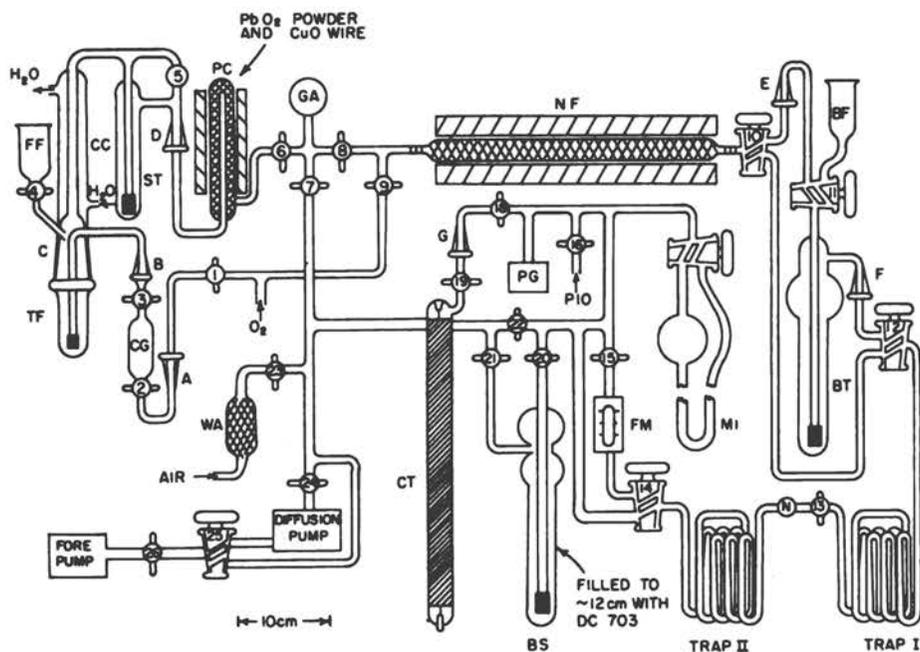

Fig. 2. Vacuum line for separating and purifying C^{11}

TF	=	target filling flask	N	=	needle valve
CC	=	condenser	FM	=	flow meter
FF	=	flask filling funnel	MT	=	calibrated manometer
ST	=	sulfuric acid scrubbing tube	BS	=	barostat
			PG	=	Phillips gauge
PC	=	PbO_2/CuO trap	CT	=	counting tube
CG	=	carrier gas cell	1-26	=	stopcocks
WA	=	water absorption tube	A-G	=	ground glass joints
GA	=	-30" to +15" Bourdon gauge		=	medium porosity scintered glass plugs
NF	=	N_1O_2 furnace			
BT	=	$Ba(OH)_2$ bubbler	BF	=	bubbler filling funnel

Procedure 7.   Production of $C^{11}O_2$ from Deuteron Irradiated B_2O_3.

Source: P. Nahinsky et al., J. Am. Chem. Soc. 64, 2299 (1942).

It was observed that the bulk of the C^{11} activity produced from
B_2O_3 irradiated with 8 Mev deuterons was expelled from the target material
and was present in the irradiation vessel as a mixture of $C^{11}O$ and $C^{11}O_2$.
The gas was swept through a CuO furnace with CO_2 carrier gas to convert the
$C^{11}O$ to $C^{11}O_2$. The $C^{11}O_2$ was then used in the synthesis of organic acids
and kinetic and structural studies were made on the labeled compounds.

NITROGEN

Procedure 1.   Yield of N^{13} from High-Energy Proton Irradiation of
Al, Zn, In, Pb, and U foils.

Source: I. Dostrovsky and J. Hudis, to be published in Phys. Rev.

It was shown that when the metal foils are dissolved as indicated
below all the N^{13} was present as NH_3. The apparatus used is shown in Fig. 3.

1. Target foils were dissolved in about 5 ml of dissolving mixture
(see below) in a 40 ml centrifuge cone. After the sample was completely
dissolved, the solution was boiled for about one minute to expel volatile
activities.

2. The solution was placed in distillation flask A, 10 ml 10M NaOH
was added through the hypodermic syringe and the solution boiled to distill
NH_3 into flask B, aided by a stream of He. Flask B contained 1N NaOH plus
$KMnO_4$ at 0°C. The stream passed through tube C containing saturated boric
acid and brom cresol green indicator at 0°C. and as long as flask B is at
0°C no NH_3 comes over.

3. Tube C is replaced by tube D containing fresh saturated boric acid
and indicator at 0°C and flask B is heated to distill NH_3 into D. It was
found convenient to have in D an amount of HCl equivalent to about an 80%
yield of the NH_4OH carrier initially added to judge the progress of the
distillation.
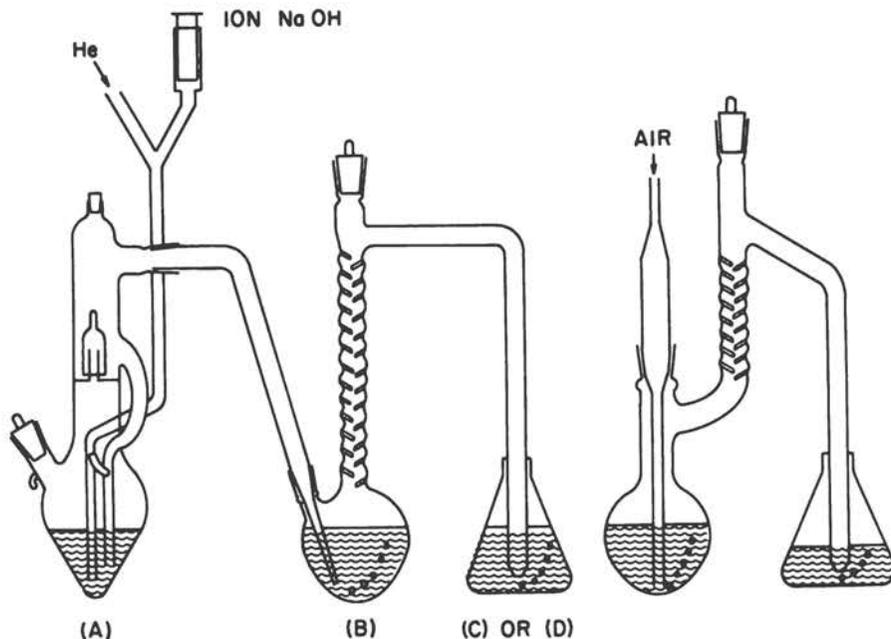
Fig. 3. Apparatus for separation and purification of $N^{13}H_3$.

4. Distillation is continued about one minute after the indicator in D has changed color and the contents of D transferred to a separate distillation apparatus.

5. 2 ml 10N NaOH were added to the solution and NH_3 distilled into a 50 ml flask containing saturated boric acid, a known amount of HCl and indicator at $0^{\circ}C$.

6. The receiving flask was stoppered, placed on top of a 2 x 2 NaI crystal and the decay of the N^{13} activity followed.

7. The chemical yield was determined after completion of the counting by titrating the solution with standardized 0.1 N HCl.

Remarks

1. Al and Zn dissolved in 6N $HCl(Fe^{++})$ plus $PtCl_4$ solution.

   In dissolved in 12N $HCl(Fe^{++})$ plus $PtCl_4$ solution.

   Pb dissolved in HBr(48%, Fe^{++}) plus $PtCl_4$ solution.

   U dissolved in 6N $HCl(Fe^{++})$.

17

2. HCl and HBr were treated with iron wire just prior to use to insure the presence of Fe^{++}, which was considered helpful in preventing oxidation of reduced forms of nitrogen.

3. The addition of $PtCl_4$ to the dissolving solution is very helpful in reducing the time required to dissolve the target foils.

4. It was observed that the presence of water greatly reduced the speed with which the Pb foils could be dissolved. In this case the desired amount of NH_4OH carrier solution was added to a few ml of HBr and the solution boiled until HBr fumes were observed.

Procedure 2. Yield of N^{13} from High-Energy Proton Irradiation of Al.

Source: K. Chackett et al., Proc. Phys. Soc. (London) 69A, 43 (1956).

This activity appears in the gas phase on dissolution of the foil in boiling NaOH solution, to which NH_3 carrier has been added. It may be trapped on a filter pad moistened with dilute sulfuric acid and counted as ammonium sulfate.

Procedure 3. Assay of N^{13} in NH_3.

Source: G. S. Golden, Combustion Engineering Inc., Nuclear Division, Windsor, Connecticut.

Reagents

NH_3 carrier	ethyl alcohol
NaOH	diethyl ether
5% chloroplatinic acid	

1. Add the sample in solution to a 125 ml Erlenmeyer flask. Add 10 mg NH_3 carrier and make basic with NaOH solution.

2. Add 2 ml chloroplatinic acid (5%) to a centrifuge cone and dilute to 10 ml with alcohol. Place in an ice bath.

3. Place a one-hole stopper on the Erlenmeyer flask. Insert a bent glass tube so that one end is above the liquid in the flask while the other is immersed in the H_2PtCl_6 solution.

4. Heat the sample to boiling and boil vigorously for about 3 minutes.

5. Filter the precipitate. Wash with a few ml of alcohol and then with ether.

6. Weigh and count.

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