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FUNDAMENTALS OF DAMAGE IN LASER GLASS

REPORT OF
THE COMMITTEE ON THE FUNDAMENTALS OF
DAMAGE IN LASER GLASS

NATIONAL MATERIALS ADVISORY BOARD
Division of Engineering - National Research Council

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Nicolaas Bloembergen
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Ad Hoc Committee on the Fundamentals of
Damage in Laser Glass

I. INTRODUCTION

Since laser action in glass was first demonstrated in 1961, the Advanced Research Projects Agency and the Department of the Navy have had a very tangible interest in the development of laser-glass materials. At the present time, military requirements exist for laser-range finders, target illuminators and designators, optical radar sources, long-range and underwater laser-imaging systems, Q-switched-laser sources, and plasma-heating devices. For range finders, target illuminators and designators, glass is a primary candidate in applications requiring 3- to 75-joule pulses. Where eye safety is of concern, erbium glass (1.5μ) currently has the edge over all other materials for both high- and low-pulse energy requirements. For most applications requiring 50- to 200-joule pulses in less than a microsecond, Nd glass is the preferred material. If high-power frequency doubling can be accomplished, glass becomes the favored material for underwater imaging.

In spite of the fact that substantial amounts of money have been expended by both industry and the military, optically induced damage in glasses still limits the usefulness of laser-glass devices. It is believed that the existing material limitations are not insurmountable. It is therefore desirable to evaluate and review the efforts in various industrial and governmental laboratories to define the fundamental material limitations and to indicate steps in research and development necessary to reach those fundamental limits.

From the point of view of efficiency, the laser should operate at, or a few times above, the saturation-energy density. For straight Nd glass this is about 10 joules/cm^2, but for Nd-Yb glass where the lifetime is 4 to 16 milliseconds, the saturation energy is greater than 40 joules/cm^2. Experience

Experience has taught systems engineers that if reliable long-term operation of a laser is desired, they should test the system a few times at approximately twice its desired output-energy density. If this test is survived, the system will very likely have a good life expectancy. Thus a glass-damage threshold of the order of 100 joules/cm^2 for Q-switched operation is a desirable goal. In addition to a higher-damage threshold, other properties such as low-loss coefficient, a high-slope efficiency, a strong resistance to solarization, an ability to obtain good yields of high-optical quality, etc., are desirable.

With this background, the Office of Naval Research (ONR) in June 1969 requested that the National Materials Advisory Board of the National Academy of Sciences-National Academy of Engineering-National Research Council initiate an appropriate committee study to explore the causes of damage in laser glass and to recommend a program directed toward a solution to the problem. More specifically, the task of such a committee was to determine what problems need to be overcome to enable our national capability to produce a laser glass having a damage threshold of 100 joules/cm^2 while still possessing the other qualities requisite for good, high durability, laser glasses.

II. THE CHARGE TO THE COMMITTEE

The National Materials Advisory Board accepted the charge and established an ad hoc Committee on the Fundamentals of Damage in Laser Glass in August 1969. The Committee addressed itself to the following guideline-type questions:

- What is the current state of the art in glass-laser materials?
- What are the fundamental scientific limitations setting an ultimate damage threshold in glassy materials?
- What are the technological limitations that, at present, prevent the ultimate threshold from being attained?
- What steps can be taken in research and development to remove present technological roadblocks to achieving the fundamental threshold limit?
- What effort in terms of manpower, time, and cost is necessary to achieve a glass-laser material with a damage threshold of 100 joules/cm^2 in a laser pulse of 10- to 100-nanosecond duration?

With this basic charter, the ad hoc Committee on the Fundamentals of Damage in Laser Glass held 6 two-day meetings during the period September 1969 - June 1970. Presentations from experts of the major laser-glass manufacturers in the United States and France, as well as from leading research experts in government and university laboratories, were heard. This report contains the results of the deliberations of the committee members, liaison representatives, and invited speakers.

III. GENERAL BACKGROUND AND ORGANIZATION OF LASER-GLASS DEVELOPMENT

When the study was started, it was believed that questions mainly of a scientific nature impeded the efforts to produce high-performance laser glass in the United States. As the study progressed, it became increasingly clear that questions of organization and technological manufacturing techniques play roles as important as the questions of materials science. The developments in the state of the art during the course of the study were significant, and it is now clear that the achievement of a laser with a damage threshold of 100 joules/cm^2 is not an isolated materials problem but will require a systems approach. The geometric design of the laser components, the balance between size of glass-building blocks and useful yield, the balance between production cost and quantity of laser glass required, all play an important role in addition to the laser-glass composition and characteristics.

The mandate to the Committee, and consequently the study, emphasizes the fundamental materials aspects of the problem. The conclusions and recommendations are restricted largely to these aspects. Although the Committee has neither the authority nor the competence to make recommendations about the modes of organization and funding of glass-laser development, the following observations are offered to provide a broader context and background against which our recommendations and conclusions must be viewed.

In foreign countries, notably France and the Soviet Union, a comprehensive and long-range development program for a particular application of high-threshold, laser-glass production was directed by a single government agency. However, the corresponding effort in the United States was supported and supervised simultaneously by a number of government agencies and private industrial institutions with diverse purposes. It is quite evident that this latter mode of operation leads to a diffuse effort that may not expeditiously solve key materials problems.

While the first glass-laser action was demonstrated in a private industrial laboratory in this country in 1961, France and Germany were capable of delivering laser glass with higher threshold for damage at the time the present study was begun. For a while the Soviet Union held the lead in the power output of Nd-glass-laser amplifiers, and the first laser-induced, thermonuclear plasma was achieved in that country. During the past year, i.e., during the course of this study, the capabilities in the United States have improved considerably. Since the state of the art is approaching the fundamental limits set by material properties and geometrical configurations, coordination of effort becomes increasingly more important. A close liaison between user and producer of laser glass at all stages of future development is desirable. Since the eventual demand for laser glass as well as further development costs are uncertain, continued funding of this development from private sources cannot be counted on.

Having pointed out that questions of organization and funding are important, the Committee will restrict itself in the remainder of this report to basic questions of materials science and technology in the production of laser glass with a high-damage threshold. The major findings of this study are summarized in the next section. The remaining sections review in more detail the state of the art and the problems presented by each of the seven subject areas into which the field could be divided. These are:

- Laser Glass Composition and Manufacturing
- Metallic Inclusions
- Nonmetallic Inclusions
- Damage Induced by Inclusions
- Surface Damage
- Intrinsic Bulk Damage
- Test Facilities

IV. SUMMARY OF CONCLUSIONS AND RECOMMENDATIONS

In this section the main conclusions and recommendations of the report are summarized. They result from the more detailed discussions in sections V to XI, which should be consulted for both supporting evidence and the general context in which the conclusions and recommendations should be viewed.

Conclusion 1

The three main causes for damage in glass-laser materials are:

a. Absorption of light by metallic and nonmetallic inclusions followed by heating and failure by thermal-stress fields around the inclusions.

b. Surface damage accompanied by plasma formation at the surface.

c. Intrinsic bulk damage in the glass, initiated by a self-focusing mechanism resulting in heating and plasma formation and filamentary-damage tracks.

Conclusion 2

The state of the art at the beginning of 1970 is such that inclusions present the most serious bottleneck in laser glass manufactured in the United States. Glass with a damage threshold of 20 joules/cm^2 in a 30-nanosecond pulse can be produced consistently with a reasonable yield in both platinum and ceramic crucibles and continuous tanks. The elimination of metallic and nonmetallic inclusions has progressed and will progress further by careful attention to technological details of the laser-glass manufacturing process. To achieve improvement it is essential to integrate the manufacturing and testing techniques. It appears that the inclusions in glass-laser rods can be eliminated to such an extent that a threshold for inclusion-induced damage of 100 joules/cm^2 in a 30-nanosecond pulse in an operating device is feasible.

Conclusion 3

The physical mechanisms for surface damage are only partially understood. Raising the surface-damage threshold by chemical and mechanical treatment of the surface to over 100 joules/cm^2 appears possible.

Conclusion 4

The onset of self-focusing with subsequent breakdown constitutes the fundamental limit to damage in bulk-laser glass, as well as in crystals. The threshold for electric breakdown and plasma formation lies over 1,000 joules/cm^2 in a 30-nanosecond pulse. Such intensities can, however, be reached by the self-focusing mechanism for laser beams with a much lower initial intensity. Although the basic physical mechanisms for this self-focusing effect are reasonably well understood, the self-focusing threshold depends sensitively on the spatial and temporal structure of the laser pulse and is influenced by inhomogeneities and inclusions. This dependence is not sufficiently well known at present. For present laser geometries, not specifically designed to avoid self-focusing damage, the threshold for the formation of self-focused filamentary-damage tracks appears to lie between 50 and 100 joules/cm^2 for a 30-nanosecond pulse. These damage mechanisms and thresholds are not very sensitive to the bulk composition.

Conclusion 5

Presently available manufacturing techniques hold promise for the production of laser-glass devices, in which the damage threshold is determined by the self-focusing mechanism. To optimize the yield of glass with acceptable loss coefficient and to optimize the system geometry for maximum self-focusing threshold, a systematic use of testing facilities and their integration with technological production processes is required. With these optimization procedures, a system with a damage threshold of 100 joules/cm^2 in a 30-nanosecond pulse appears feasible.

Conclusion 6

To achieve a glass-laser system with a damage threshold of 100 joules/cm^2 in a 30-nanosecond pulse, a concerted effort on all causes of damage listed above will have to be made.

Conclusion 7

Much of the experience of neodymium laser glass may be transferred to the development of erbium laser glass. Some new problems may arise if a base other than a silicate is required.

Recommendations

As a result of these above conclusions, the following recommendations are listed in their approximate order of priority. However, in view of Conclusion 6, it is felt desirable that all those recommendations listed from 1 through 5 be implemented. All man-years of effort listed in all the recommendations should be carried out over a 1- to 2-year time period. (For example, where an effort of 4 man-years is specified, this means 4 men for 1 year or 2 men for 2 years.)

Recommendation 1

The development and further improvement of laser glass manufactured in refractory ceramics should be continued. High priority should be given to the identification of the damage sites and to the fundamental study of their origin. The influence of various ceramic materials on the damaging inclusions and absorption coefficients of laser glass should be studied. The identification of damage sites in glass with acceptable low absorption and small-quantity production work with improved ceramics is expected to overlap.

A minimum effort of 5 man-years is necessary. A healthy effort of 7 man-years is recommended. The cost for this program is expected to lie between $300,000 and $400,000.

Recommendation 2

The program to produce laser glass in platinum containers should likewise be continued. Emphasis should be placed on obtaining thermodynamic data for the oxides contained in laser glass and for metal bases in platinum. If these data confirm the feasibility of producing an inclusion-free, laser glass in platinum at theoretically predicted optimum-oxygen pressures, further experiments to confirm the improved properties of glass produced under these optimum conditions should be carried out.

A 3 man-year effort is necessary to obtain the basic thermodynamic data at a cost of about $200,000. A 2 man-year effort on mapping and analysis would cost about $75,000. Subsequently, an additional 2 man-year effort for producing glass under optimum conditions at a cost of $150,000 may be warranted.

Recommendation 3

A systematic study of surface damage of glasses should have high priority since the surface-damage threshold is often lower than the objective of a 100 joules/cm^2 limit in a 30-nanosecond pulse and the damage mechanism is not fully understood. Systematic data on damage threshold should be obtained of glass samples with controlled surfaces. (The measurements should include fresh outgased samples with surfaces in vacuum, surfaces with controlled amounts of known adsorbed gases, samples with mechanically hardened surfaces, glass surfaces immersed in a liquid that may be matched in optical index of refraction or in shock-wave impedance. The dependence of surface damage on temperature, spot size, and pulse duration should be investigated. Systematic studies of the influence of etching and aging in the atmosphere should also be conducted. Scanning electron microscopy of surface-damage sites is recommended.) This program should be integrated with the manufacturing process of glass-laser elements and with the associated test programs for inclusions and bulk damage.

A minimum effort of 3 man-years is considered necessary. The cost is estimated at $200,000.

Recommendation 4

Since the fundamental limit for filamentary-track damage determined by the onset of self-focusing has already been reached in a few selected configurations, further theoretical and experimental studies of self-focusing and mode distortion as a function of the temporal and spatial structure of the beam and the geometry of the output elements of the Q-switched laser are recommended. The study of self-focusing in the picosecond-pulse regime should also be continued. Raising the fundamental threshold by a change of the material properties of bulk glass, including the elastic moduli and optical index, should be pursued with lower priority because the maximum factor for improvement, obtainable by extensive and expensive glass-materials engineering, is not expected to exceed a factor of 2.

An effort of 4 man-years is recommended. A minimal effort would include 2 man-years. Existing test facilities should be used. The estimated cost of this program for the recommended effort of 4 man-years is $200,000.

Recommendation 5

It is recommended that existing programs of test facilities, both in governmental and industrial laboratories, be continued. These test facilities are essential for the implementation of the other recommendations. Consequently, their maintenance and further development should be given a high priority. Full advantage should be taken of the existence of special capabilities and the complementary nature of different installations.

The manpower requirements for damage testing of laser glass lie between 1 and 2 man-years per test facility per year. The annual cost per test facility is estimated to lie between $50,000 and $100,000. Part of the cost of the test facilities in industrial laboratories would be absorbed in the estimated requirements of the other recommendations.

Recommendation 6

Whereas Recommendations 1 and 2 can be carried out with crucibles of modest size at lower cost, the continuous tank-melting process has many advantages and should be adopted, if the demand for larger quantities of laser glass and the feasibility of producing laser glass with the required specifications in crucibles have been demonstrated.

The estimated minimum cost for a ceramic tank facility is $750,000.

Recommendation 7

Close attention should be paid to research results in eye-safe wavelength lasers in glass hosts so that timely establishment of test facilities can be made.

The estimated cost for an erbium glass-test facility is $200,000.

V. LASER-GLASS COMPOSITIONS AND MANUFACTURING

General

Neodymium-glass lasers have been made in a variety of hosts, including silicates, phosphates, germanates, lanthanum borates, and aluminates.(1) The principal requirement for a good host is that fluorescence of Nd^{3+} occur without quenching and that the glass be transparent at the laser wavelength of 1.06 microns and at the pumping bands in the visible and near infrared. In addition, the glass should be capable of being fabricated in good optical quality in large samples. The glass should also be free of solarization, or if there is some discoloration on exposure to pumplight, efficient operation should also be possible with filters that remove the damaging ultraviolet light. The final requirement, and the one which is of primary concern in this report, is that the glass also be free from damage when exposed to the high-power levels of Q-switched operation.

The particular choice made by different glass manufacturers, or laboratories working with laser glass, was often based on peripheral considerations such as the experience gained with a particular glass system in connection with other glass fabricating activities, or the availability of relatively "clean" materials from which the glass could be made. These factors have led all of the principal glass manufacturers to work with alkali, alkaline-earth silicates because they have had considerable experience in fabricating these glasses and can readily make good optical quality material in large sizes. Furthermore, the starting materials from which silicate glasses are made are relatively free of contaminating colorants, particularly iron and copper. The transition-metal ions that are most serious in producing absorption at 1.06 microns are Ni^{2+}, Co^{2+}, Cu^{2+}, Fe^{2+}, and V^{3+}.(2) These practical considerations have made alkali, alkaline earth, silicate glasses a commonly fabricated laser glass even though these glasses have problems of solarization and have fluorescent linewidths that are broader than is often desirable for many applications. Some phosphate-host glasses can be made free of solarization and have narrower linewidths thereby permitting lower thresholds for some applications. However, the difficulty in making large volumes of phosphate glasses with good optical quality and the problem of avoiding contamination due to iron and other transition-metal ions have thus far obviated any practical role for them.

The starting materials for fabricating alkali, alkaline-earth silicates usually consist of silica in the form of a finely divided quartz mixed with the other ingredients, one or more of which are not oxides. Carbonates, hydroxides, nitrates are commonly used for the alkali, alkaline earth, or aluminum, if it is present, in order to "flux" the original batch. In the melting of the glass, the mixed oxide is formed with the evolution of gas, some of which initially appears as bubbles within the glass. Some glasses have sufficiently reactive constituents so that it is possible to use oxides for all the starting materials. The next stage in processing is a refining or "fining" step in which bubbles are cleared out of the glass. Fining agents are often included in the glass which, at an appropriate temperature, cause an enlargement of the bubbles so that they readily rise to the surface and leave the melt. The effectiveness of a fining agent depends upon the glass. Commonly used agents are the oxides of arsenic, antimony, and cerium. In addition, the alkali chlorides are sometimes suitable or, for some glasses, none are required.

After the glass has been fined, the next step is homogenization in which the glass is stirred to provide good optical quality, striae-free glass. The glass is now ready to be poured or tapped from the furnace or, in the case of the older "transfer" process, is left to cool in the crucible.

Prior to World War II, it was customary to use ceramic crucibles in most glass manufacturing plants. However, since then, platinum crucibles and platinum tanks came into wide use because platinum is not readily attacked by the silicate glass thereby allowing much more rapid and more effective homogenization. The very limited crucible attack permitted much longer life. Furthermore, with the reclaim value for the platinum crucible, the cost for making glass in platinum is less than in some refractories.

In continuous tanks, with platinum in the stirring section, the batch is loaded at one end and finished glass emerges from the other end. It is not uncommon that the first stage is made of ceramic materials for the glass forming and refining reactions. This is done because often the unreacted batch is chemically extremely active and the platinum could be attacked. Even if some ceramic attack does occur in the reacting and refining stages of glass fabrication, this is not often a problem because the homogenizing still occurs in platinum so that whatever refractory does dissolve is homogeneously mixed within the finished glass.

While platinum crucibles are relatively inert, some pot attack can take place. There are four principal ways in which this can occur. Physical abrasion and corrosion can lead to particles of platinum flaking off into the melt. In an oxidizing atmosphere, platinum-oxide vapor is formed, followed by condensation on portions of the furnace or even the melt surface, and then subsequent reduction to the platinum metal within the glass. Finally, in an oxidizing atmosphere, platinum could be dissolved directly into the glass from the pot, principally at the glass-air interface. With proper handling in the preparation of the pot and the positioning of the stirring rod, abrasion should be avoidable. The corrosion could be kept within acceptable limits by frequent reworking of the platinum. The other two methods of platinum contamination in principle could be coped with by preparation of the glass in a neutral or slightly reducing atmosphere. A strongly reducing atmosphere can cause problems with reduction of other constituents which, in turn, could form an alloy with the platinum thereby leading to excessive pot attack.

In addition to platinum-pot attack, another source is the small amount of platinum in the batch which could produce serious problems from the point of view of inclusion damage unless it is dissolved. To dissolve the platinum requires an oxidizing atmosphere at least in the initial stages of melting.

In the case of a ceramic crucible, one of the major problems is the elimination of striae. It is necessary to find a combination of pot material and glass composition which gives only limited pot attack, and to have a time-temperature cycle and stirring such as to effectively homogenize the glass. The pot attack can lead to a higher loss coefficient if the ceramic is "dirty," i.e., contains too high a concentration of transition-metal ions. By a proper selection of glass composition and ceramic-crucible material together with appropriate time, temperature, and stirring cycles, it is possible to obtain striae-free, low-loss glass. Even though there are no known sources of platinum present, damage sites can occur due to as yet unknown causes.

A variety of test procedures have been used to evaluate the glasses of different manufacturers. It is important to stress that the presence of inclusions is inherently a statistical problem. The analysis presented by Uhlmann(9) indicates that the threshold for damage is a function of the particle size and duration of the pulse; hence, in a given glass sample the most complete characterization of inclusion damage is the number per-unit volume of damage sites as a function of incident-energy density for various pulse lengths. Unless the glass has so low a density as to be virtually free of inclusions, and short of a complete description of the number of sites versus energy and pulse duration, the most meaningful data in characterizing a given glass sample is the concentration of damaging inclusions as a function of energy density for a particular test procedure. This still leaves open the comparison of one test procedure with another. The scale factor to use in comparing the energy density for damage for one pulse duration to that in a different pulse time would have to be determined experimentally for a particular glass.

Because of the statistical nature of the damage sites in glass, it is necessary to irradiate large volumes with known energy densities. In active tests, only the last few centimeters of path length in an oscillator-amplifier system are subjected to energy densities comparable to the output value. Hence, active tests usually provide data on relatively small volumes of glass. In passive tests, long rods may be used but care should be taken to avoid self-focusing in this case. For a given geometry and total pulse energy, it is necessary to optimize the pulse duration to discriminate between inclusion and self-focusing damage.

If the density of damage sites is sufficiently low (less than 1 per liter), or if it is very high (in excess of 1 per cubic centimeter), the statistics of damage sites are less important in evaluating the glass in question.

Programs of Principal Laser Glass Manufacturers

American Optical Corporation

The original work on laser glasses at American Optical (AO) was in platinum crucibles. After it was recognized that the principal sources of laser-glass damage were platinum inclusions, AO's first attempt at damage-resistant glass was in a platinum crucible in a controlled atmosphere.(3,4,5) To dissolve any platinum that might be present in the batch and to preclude excessive pot attack by the active batch constituents, the glass was first reacted and partially fined in a high-purity mullite crucible. It was then transferred to the platinum crucible in a neutral atmosphere for the remaining glass processing.

After erratic results with platinum crucibles in an atmosphere that could be controlled to be either neutral, slightly reducing or slightly oxidizing, AO went to an all-ceramic system made of high-purity mullite with a capacity of 23 kg of glass in the crucible and about 16 kg of extruded output in the form of 7.5-cm-diameter billets by 1 meter in length. These billets were tested with 80 joules/cm^2 in a pulse that had a 4-μsec, full-width at ½ intensity but a total duration of 10 μsec. Only glass that survived this test was considered suitable for high-intensity, Q-switched operation at energies up to about 20 joules/cm^2 in a 30-nanosecond pulse. The composition of the glass that has been manufactured and tested in sufficient quantity to provide significant damage and yield data is given in Table I.

Prior to damage testing, the only visible inclusions were occasional bubbles. The yield of glass free of striae and with a sufficiently low density of bubbles to be useful in a laser device is about 35 percent. The density of damage sites that appeared on testing with the above-mentioned pulse averaged one damaging inclusion per liter. The yield following the damage test depends on the rod size required. For small rods, 6-mm diameter by 7-cm long, very little glass is lost, but for large rods, 25-mm diameter by 50 cm, the yield is about 60 percent.

In passive tests on recently prepared experimental glass, with the amplified spontaneous emission device operated to give a pulse duration of 120 nanoseconds, no damage was observed at 100 joules/cm^2 in a sample volume of 20-cubic centimeters.

Owens-Illinois, Inc.

The glass presently being made by Owens-Illinois (O-I) is made in a platinum crucible under controlled atmosphere conditions.$^{(6)}$ Its composition and some data on yield, loss coefficient, and damage are given in Table I. In addition to controlling the atmosphere at an appropriately low-oxygen partial pressure, there is some benefit from the glass composition because it is prepared at temperatures somewhat less than other commonly used laser glasses. Other things being equal, the reduced temperature inhibits platinum-pot attack.

The laser glass manufactured by Owens-Illinois is inspected with a visible light source by a trained operator who can detect particles of a size as small as 1 micron. Only glass with a particle density less than approximately 5 particles per liter is accepted for subsequent fabrication into laser rods. The yield of glass at this step is approximately 15 to 20 percent. Care is taken to avoid any visible inclusions in fabricating rods from this selected glass.

Passive damage tests on selected small samples, 4-mm thick by 2-cm diameter, of Owens-Illinois laser glass have not shown damage at 70 joules/cm^2 in a 55-nanosecond pulse when irradiated with the nonfocused output from the Brewster-angle end of the Owens-Illinois oscillator-amplifier system.(7)

Active tests in which Owens-Illinois laser glass is the last stage of amplification in a Q-switched, oscillator-amplifier have indicated damage seldom occurs up to 20 joules/cm^2 in a 55-nanosecond pulse for laser rods 19-mm diameter by 21-cm long. Rods tested up to 40 joules/cm^2 can show some inclusion damage but the inclusions are small enough so that no further degradation occurs after the first pulse on repeated exposure of up to 200 shots at 40 joules/cm^2. Selected samples have withstood up to 50 joules/cm^2 in a 55-nanosecond pulse. Yield figures on the damage testing at more than 20 joules/cm^2 at 55 nanoseconds are not yet available, nor is data on the density of damage sites as a function of energy density.

Sovirel (France)

The first laser glass melted by Sovirel in France had the composition shown in Table I and was melted in a platinum crucible in a semicontinuous process.(8) This showed inclusion damage due to the presence of small particles of platinum for energies of 7 joules/cm^2 in a 30-nanosecond pulse. Their effort was then divided between work with a controlled atmosphere using a platinum pot and an all-ceramic system.

Sovirel's experience with a platinum crucible in a controlled atmosphere gave erratic results similar to those found by AO.

Parallel with Sovirel's effort in producing glass in a platinum crucible with a controlled atmosphere, work was undertaken with an all-ceramic system. A number of melts were made in alumina crucibles in pot sizes of 1, 5, and 30 liters. They found that the alumina crucibles were a distinct improvement over the platinum with energy-damage thresholds in the neighborhood of 25 to 30 joules in a 30-nanosecond pulse, and in one case as high as 54 joules/cm^2; however, inclusion damage was still present.

Sovirel's next effort was to use large clay refractories that were traditional before World War II in the glassmaking industry. Glass produced by this method did not contain any visible inclusions and consistently withstood damage thresholds of 40 to 50 joules/cm^2 in a 30-nanosecond pulse. When the glass damaged at this value or higher, it was due to self-tracking. The disadvantages of this process were the relatively low yield (typically only 20 percent) and the relatively high-loss coefficient at the laser wavelength (½ to 1 percent/cm loss). Furthermore, this procedure for making glass required considerable skill on the part of the glassmakers and is a process that is being discontinued. This is one of the laser glasses that is now available commercially and which is used in the Q-switched, oscillator-amplifier device manufactured by the Compagnie Generale d'Electricitie (CGE, France).

The most recent laser glass made by Sovirel was made in a ceramic-continuous tank fabricated from a cast refractory (ZAC 1681, made by Electro Refractories). This glass has the low-loss coefficient of ¼ percent/cm, and the high yield of 85 percent of striae-free, bubble-free glass. Damage first appears at an energy density of 22 joules/cm^2 in a 30-nanosecond pulse. The damage sites have a density of 300/liter. At higher-energy densities, no new damage sites appear but those that showed up at 22 joules/cm^2 tend to grow in size to several mm. Work is presently being carried out to identify the damage sites. They cannot be seen prior to damage and, at present, are of unknown character and origin.

Schott Glass Company (Germany)

Up until the summer of 1969, the laser glass made by the Schott Glass Company was produced in large clay pots. The glass was free of damaging inclusions but had the relatively high-loss coefficient of ½ to 1 percent/cm. Damage due to self-tracking occurs at energy densities in excess of 50 joules/cm^2 in a 30-nanosecond pulse.

Erbium Glasses

In addition to Nd glasses that lase at 1.06 microns, there is an interest in erbium-glass lasers that emit in the eye-safe region of 1.54 microns. Test samples of Er-laser glass have been made by American Optical in the same silicate base (exclusive of rare earth) used for their Nd laser and in a Zn-Al-phosphate. Owens-Illinois has made Er-laser glass in their Li-Ca-Al-silicate. If the silicates prove to be most suitable for Er glass, as they have for Nd glass, it is expected that the same base glass and melting facility would be used as required for Nd. Because of the three-level character of the emission, concentrations less than 0.5-wt. percent of Er are used. For efficient operation at room temperature, Yb_2O_3 is added as a sensitizing agent in concentrations in excess of 10-wt. percent, and in some glasses small amounts of Nd_2O_3 (< 0.2-wt. percent) are also added for further sensitization. The differences between the Nd lasers at 1.06 microns and Er lasers are the obvious differences in wavelength, the rare earths used, and the total rare-earth content. The wavelength should not have any effect on the inclusion-damage problem, but the different rare earths and particularly the higher rare-earth content could have an effect insofar as the melting properties of the glasses are altered. Glasses melted in platinum should not be radically changed, but until the sources of inclusion damage in ceramic melts are known the situation is unclear.

Summary and Recommendations

Unless the density of damaging inclusions is reduced to a negligibly small value, the problem of damage sites in laser glass is essentially a statistical one. A complete characterization of a glass sample would include the number of damage sites per-unit volume as a function of energy density for various pulse durations. These data are not yet available for low-loss laser glass manufactured in the United States. However, glass manufactured by both American Optical and Owens-Illinois is sufficiently low in damaging inclusions to permit reliable damage-free use in laser devices up to energy densities of 20 joules/cm^2 in a 30-nanosecond pulse.

The experience of both AO and Sovirel with platinum melters in a controlled atmosphere was not encouraging enough for either company to pursue the method. Owens-Illinois feels that by its present methods of quality control, which include both inspection for large particles and damage testing under conditions comparable to those employed by the user for elimination of excessive numbers of small-damage sites, useful glass that does not deteriorate beyond the damage shown after the initial irradiation can be obtained for energy densities up to 40 joules/cm^2 in a 55-nanosecond pulse for rod sizes 25 mm in diameter and 30 cm or more in length. The ability to eliminate inclusions due to platinum with a platinum crucible in a controlled atmosphere will depend in part on obtaining the necessary thermodynamic data for the oxides in laser glass and for metal and oxide solutions in platinum.

By use of 300-liter-clay pots, both Sovirel and Schott have demonstrated that laser glass can be produced with negligible concentrations of damaging inclusions; however, the naturally occurring clay materials used for the crucible give absorption losses that, for many applications, are unacceptably high. American Optical and Sovirel have demonstrated that by the use of synthetic ceramic-crucible materials, low-loss, damage-free glass can be produced for energy densities up to 20 joules/cm^2 in a 30-nanosecond pulse. American Optical used the 8-liter-mullite crucibles with the yield of 35 percent, and Sovirel a continuous tank made of ZAC 1681 refractory with the yield of 85 percent. It is probably necessary to use synthetic ceramic-crucible materials to avoid absorption due to excessive concentrations of transition-metal ions and thus obtain low-loss glass. To reduce the density of damaging inclusions to acceptably low values, is a problem that is common to both types of melt facilities. The decision as to which facility ought to be used depends upon the anticipated quantity of laser glass required both in total volume and in rod sizes needed. The 8-liter-batch process is inherently less efficient but the initial cost for installation is considerably less. If national requirements call for large volumes of glass, the efficiency for production in continuous tanks could more than offset the initial cost. It should be emphasized, however, that a further reduction in damaging inclusions resulting from the use of ceramic-melt facilities can come about only by further work on the identification of the inclusion and elimination by proper selection and/or treatment of the crucible material.

The analysis of small inclusions is difficult because the included material after damage is spread over a large volume. A nondestructive test that would identify the position of the inclusion could help in the use of microanalysis such as electron-beam probe, spectral analysis with a micro-laser-beam probe, or whatever other method is used. With the identification of the inclusion, hopefully, it could then be eliminated.

Suggested Programs and Cost Estimates

The recommended research to obtain glass free of damaging microinhomogeneities by use of a ceramic-melt facility is listed below. In addition, a program to test Er lasers at 1.54μ is also given.

1. To help in the elimination of damaging inclusions in the presently made laser glass and to provide a method for coping with such problems in the future should they arise, a nondestructive method to locate the position of the site would be of great value. This could be done by a holographic method. Then micro-probe analysis would be used to identify the composition of the site. This program could also be of value to identify the composition of inclusion in glass melted in a platinum facility. The work would be carried out in parallel with a program to manufacture the glass.

No new capital equipment beyond that at present available to the glass manufacturers would be needed. A 3 man-year effort would be required at a cost of $200,000.

2. If the need for a large volume of laser glass is likely to warrant it, an all-ceramic, continuous-tank facility offers excellent promise to produce low-loss, damage-free glass with high yield. Before selecting a ceramic for the tank, it is necessary to do further testing of various ceramic materials, and heat or chemical treatments of these ceramic materials to prevent their contamination of the glass with damaging inclusions. Since a

continuous tank is inherently a high-volume operation, a typical run of a few days produces about a ton of glass at a cost of approximately $25,000. The number of runs that would be required to establish all the conditions necessary for reliable production is uncertain but may be as high as 10.

The cost of the program is estimated to be a minimum of $500,000 for capital equipment plus 4 man-years of effort or a total cost of $750,000.

3. If silicate glasses are used for Er lasers at 1.54μ, it is expected that the same melt facilities and approximately the same host-glass compositions would be used as those required for Nd glasses. However, sufficient volumes of suitable high-quality Er glass would have to be produced and a test facility at the Er wavelength constructed to adequately test this glass. Following the results of this test program, an estimate can be made of the required melt facility.

The capital equipment cost for the test facility would be about $80,000. A 2 man-year effort would be required. The total cost of the program would be $200,000.

TABLE I

Nd LASER GLASS MADE BY PRINCIPAL MANUFACTURERS

Company	Composition (wt. percent)	Melt Facility	Yield* (percent)	Loss Coeff. at 1.06 microns (percent/cm)	Reported Damage Characteristics**
American Optical Corp.	SiO_2 68, Al_2O_3 2, Sb_2O_3 1, BaO 5, ZnO 2, Li_2O 1, Na_2O 7, K_2O 11, Nd_2O_3 3	8 liter mullite crucible	35 percent	0.3	No visible particles. 80 joules/cm^2 in 4-nanosecond pulse gives 1 damage site/liter (equivalent to 20 joules per cm^2 in 30 nanoseconds
Owens-Illinois	SiO_2 63, Nd_2O_3 3, Al_2O_3 6, CaO 11, Li_2O 15, CeO_2 0.5	8 liter Pt crucible in neutral atmosphere	20 percent	0.3	Quality control accepted only glass with fewer than 5 visible platinum inclusions per liter. Accepted glass has no damage up to 40 joules/cm^2 in 55-nanosecond pulse.
Sovirel (France)	I. SiO_2 64, Na_2O 6, K_2O 18, BaO 3.5, Al_2O_3 2, ZnO 2, Nd_2O_3 3, Li_2O 1, Sb_2O_3 0.5	330 liter clay pot (lip poured)	20 percent	0.5 to 1.0	Negligible particle damage. Limited by self-tracking damage ($>$50 joules/cm^2 in 30 nanoseconds.
"	II. Same composition as I (above)	Continuous tank made from ZAC 1681 refractory	85 percent	0.3	No visible particles. No damage up to 22 joules/cm^2 in 30 nanoseconds, but at this energy and higher get 300 damage sites/liter.
Schott (Germany)	Mixed alkali, barium silicate similar to AO and Sovirel.	300 liter clay pot (lip poured)	20 percent	0.5 to 1.0	Negligible particle damage. Limited by self-tracking damage ($>$50 joules/cm^2 in 30 nanoseconds

*The yield figures quoted here are for quality control in relation to visible inclusions or striae, but prior to damage testing.

**See text for further data on damage test results.

VI. METALLIC INCLUSIONS

Introduction

Metallic inclusions in laser glasses can originate from a number of sources and can include most metals; however, if one considers the specific heat, thermal conductivity, melting point, and the absorption coefficient in the region of the wavelength of laser-glass output, most metals that could occur as metallic inclusions in laser glass are seen to be quite similar in their capability for causing failure. Further, although on some occasions other metallic inclusions have been observed, noble metals, which are usually platinum or platinum alloys, are the most frequently observed metallic inclusions. The case for considering platinum as the major constituent in metallic inclusions in laser glasses is further strengthened by the fact that many high-quality glasses are either melted in platinum crucibles or have hot platinum in the vicinity of the molten glass. Of course, the resulting inclusions might be an alloy of platinum and it certainly should be possible to produce other metallic inclusions but, as stated earlier, platinum inclusions have been the most frequently observed.

Sources

Platinum can, and does, end up in glass as discrete particles and the sources are varied. The oxidation, vapor transport of the oxide, and reduction and precipitation of the metal in, or on, the glass only requires platinum and oxygen at an appropriate temperature and in the vicinity of the glass during melting. Platinum particles may fall into glass melts from crucible walls, various tubes or rods used in the hot furnace, or from homogenizing stirrers. The sloughing of crystallites from platinum-crucible walls or stirrers is another potential source. Most of these sources assume that the glass is being melted in a platinum or platinum-alloy crucible and/or there is platinum in the vicinity.

Although the addition of platinum as an impurity in the raw material batch or the dropping or sloughing of particles directly into the melt cannot be overlooked as a source of metallic inclusions, the vapor transport of platinum oxide to the melt appears to be a more significant source. The observation and recording of platinum inclusions in laser glass has taken place and, in many cases, it appears that the particles look as though they have grown in, or on, the melt or glass. That is, often the particles have well-defined crystalline faces, angles, and edges and do not have any of the eroded or damaged areas that are typical of crystalline particles that have been added to the melt rather than grown from it. These factors suggest that for glasses melted in platinum-alloy crucibles or in the pressence of hot-platinum alloys, the transport of platinum to the melt by platinum oxide and a reduction of the oxide to the metal may be the prime source of platinum-metal inclusions in the glass. However, one should not ignore the possibility that discrete platinum particles may come from platinum impurities in the raw material.

Problem Solution

Some limited thermodynamic data exist on the relationship between oxygen and platinum at elevated temperatures.(10) The data suggest that platinum inclusions in laser glass could be caused by the migration of platinum as platinum oxide. Gas-phase transport is possible with platinum dioxide gas as the intermediate species, but the transport of platinum-oxide complex through the glass does not appear to be as likely.

The obvious solution to the platinum oxide problem is to reduce the oxygen-partial pressure and thereby inhibit its formation at the melting temperatures; however, the constituents of the glass are oxides of metals with definite and definable oxide-metal-oxygen relationships. The problem then becomes one of adjusting the oxygen-partial pressure to as low a value as possible without reducing the laser-glass-oxide constituents to the metallic state. This can be accomplished by empirical experimental approaches, but it would be very time-consuming if it were done as a function of the composition of the laser glass. A better approach would be the application of thermodynamic data to the particular system. Unfortunately, the existing data do not take into account the fact that the oxides are in a glass solution and the metals (from the laser-glass oxides) are in a platinum alloy.

Obviously, the thermodynamic-activity coefficients of the oxides in the glass as a function of the composition would be very useful, but only if the corresponding-activity coefficients for the metals of the glass in platinum were also known. With this type of information, the quantity of platinum or platinum oxide that was transported to the melt could be controlled by controlling the oxygen-partial pressure while being assured that no competing reaction involving the reduced oxides of the laser glass and the platinum metal would occur.

The damage mechanism associated with a metal (platinum) inclusion is not completely elucidated, but it is assumed that the high absorption of the laser energy by the particle and the low-thermal conductivity of the glass lead to high stresses in the area of the particle. These stresses are probably a result of the differential-thermal expansion between the metal and the glass. This suggests that there is a relationship between damage (at some given energy level) and the size of the particle. Work is currently in progress in several laboratories to examine the relationship between particle size and damage threshold, both from a theoretical and from an experimental point of view. These data will be very helpful as there are very little, if any, in existence today. Data on the relationship of particle shape and stress distribution will be needed but are probably easily obtainable. Certainly, the basic room-temperature data on the properties of the glass are available for most, if not all, laser glasses. Young's modulus, Poisson's ratio, thermal expansion and diffusivity, and viscosity data at room temperature and at temperatures in excess of 1,000°C, as well as the absorption as a function of wavelength and temperature, will be needed for any calculations.

The experimental part of a program to evaluate the size, shape, and type of damage caused by metal (platinum or its alloys) inclusions will necessarily include the establishment of the location of the particle before and after it causes damaging under lasing conditions, and an analysis of the particle (before and after) to establish its composition. Work on mapping techniques is just beginning. Microscopy has had some limited success, but holography appears to hold the most promise for ease of mapping. Scanning and probe-electron microscopy are being employed to identify the particle and, although the techniques will undoubtedly be improved upon, they will probably be taxed beyond their limits of detection.

The specification of the platinum crucible from the standpoint of grain size, impurity level, and surface-to-volume ratio has received considerable attention but must be a part of any experimental or commercial program. The analysis and identification of noble-metal impurities in the raw-batch constituents cannot be overlooked regardless of their source. Most organizations that make laser-glass melts in platinum crucibles have considered the crucible and raw-materials specifications and further experimental work is probably not needed.

Recommendations and Conclusions

The necessary research to allow laser glass with a high-damage threshold to be produced without metallic inclusions is suggested below. The assumption that was made earlier is continued, i.e., metallic inclusions are platinum or platinum alloys.

1. A program to obtain thermodynamic data for the oxides in laser glass and for the metals of the oxides in platinum.

2. Experimental verification should be carried out based on (1) above to establish the usefulness of melting laser glass in platinum at the theoretically predicted oxygen pressure.

3. Mapping, location, and analysis of metallic inclusions in laser glasses should be carried out and correlated with laser-damage studies that establish size and damage-threshold relationships. This program has begun and should be continued.

Funding in terms of manpower and dollars is estimated on the basis of each of the three programs but there should be considerable overlap and parallel programming.

1. A 3 man-year effort would probably provide the basic data but an additional 2 man-years would undoubtedly be desirable. Materials, labor, and equipment, would probably cost about $200,000 for the basic data.

2. About 1 man-year effort should be sufficient for verifying the predictions of the thermodynamic data if certain necessary melting and atmosphere control facilities are available. Assuming no capital expenditures, the program could probably be carried out for about $50,000.

3. Approximately 2 man-years of effort will probably be needed to map, locate, and analyze particles if it is assumed that the necessary analytical tools already exist. This would probably cost about $75,000.

Continuing or new work resulting from the above might total another $100,000. If significant pieces of equipment are not available for carrying out the work and must be built, purchased, or rented, the cost of each program would have to be funded at a correspondingly higher level.

VII. NONMETALLIC INCLUSIONS

Introduction

There are problems with the quality of present laser glass even when it is good by conventional optical glass standards. One of the most severe of these problems is inclusions. The difficulty of controlling inclusions can be seen from experience - particles no bigger than 1 micron cause debilitating damage.(11) Since total volumes of glass of the order of $10^3/cm^3$ are used, the inclusion concentration should be below 10^{-15}. Even for small volume lasers of 1 cm^3, the concentration should be below 10^{-12}. Control of contaminant in this range is presently impossible. The only workable answer is to keep any contaminants dissolved. Concentrations of significant impurities on the order of 10^{-6} seem feasible today and almost all nonmetallics in this range are completely soluble in oxide glasses. This means that nonmetallic inclusions normally will have been introduced as some type of aggregate and will have remained so throughout the melting process because the rate of their solution was too slow. The problem is to eliminate these unknown aggregates at their origin or to dissolve them.

There are two dimensions to inclusion studies. The first is the problem of large inclusions (over about 1 micron in size). These may be detected optically and isolated for study. The bulk of present knowledge stems from such investigations. Identification of the nature of the inclusion by X-ray, electron microprobe, etc., suggests its origin and leads to its elimination. The second category of the inclusion problem involves those below about 1 micron in size. These are not easily detected and there is a serious need for suitable study techniques to be developed. Until such techniques are devised, the most profitable avenue will be studies of large inclusions, assuming that the small ones have similar physical properties and similar origins.

In the next section we discuss the origin of inclusions, in general, and their prevention with specific thoughts on which origins appear most troublesome. In the section following this, various physical weakening effects of nonmetallic inclusions are treated. The most important of these will be selected to help delineate the most serious types of inclusion. Recommendations then arise from this reasoning.

Origin of Nonmetallic Inclusions and Their Prevention

Noncrystalline Inclusions

There are two types of nonmetallic inclusion - glassy and gaseous (bubbles). Gaseous inclusions usually arise from gas occluded or exsolved during the melting process. These inclusions have been studied extensively because of their detrimental properties in many commercial applications. However, they rarely cause damage in lasers and will not be discussed further here.

Glassy inclusions arise from emulsion formation and melting of crystals without subsequent complete solution. The frozen stresses around glassy inclusions may be as large as around crystalline inclusions but the completion of the melting means that flaws will probably be fewer and they will probably be stronger. As shown below, the main detrimental property of such inclusions is their optical absorption. Most glassy inclusions, being transparent, will not cause damage. The probable high strength of such inclusions further guarantees that only relatively high-optical absorptions are important. Since absorbing ions well-dissolved will be in low concentration, emulsion formation with these as a major constituent will not occur. There is the possibility that the lasing ion may occur as an absorbing, damaging inclusion if there is a local, very large concentration. But examination of this case indicates that it is not very likely. This leads to the conclusion that the major damaging glassy inclusions will be those in which a crystal impurity melted but did not dissolve. Thus, the origin and prevention of these is covered under crystalline inclusions below.

Crystalline Inclusions

Crystalline inclusions arise from three sources: unmelted batch, devitrification, and refractory attack.(12) These will be discussed in that order.

Unmelted batch occurs from such things as grains too large, agglomeration, etc. Assuring that the batch is finely divided and well mixed normally will alleviate this problem. This can be done without difficulty for laser glass although contamination of the batch may be troublesome. Of particular importance is the presence of absorbing impurities in localized concentration much higher than the average impurity concentration (crystals of iron oxide, etc.). These might pass through the melting process without melting or with melting and incomplete solution. In either case, they present potential-damage sites. The straightforward answer is to remove such impurity concentrations from the batch or enhance the melting so that complete solution occurs. This does not appear to be a formidable problem.

Devitrification usually will not increase impurity concentration since the nucleation rate for impurity concentrations normally will be too low for crystallization to occur. Devitrification resulting in large crystals with many flaws may be troublesome on occasion, but this is straightforwardly handled by keeping all parts of the melt above the glass liquidus. Devitrification does not appear to be a formidable problem either.

Refractory attack is the most likely source of damaging inclusions. Not only are these materials normally very impure, but the impurities may occur in high concentration at grain boundaries or segregated in the original refractory raw material. Thus, they can be swept into the melt as aggregates and not dispersed readily. Furthermore, in a ceramic melter, this may occur in the later stages of melting thereby preventing solution.

Assessing this, it is concluded that highly concentrated impurity regions are the most troublesome and these are most likely to result from refractory attack.

Physical Weakening Effects

Diffraction and Lens Effects

Any high-refractive-index inclusion would tend to focus the light in a rather imperfect way and diffraction from small inclusions might similarly raise the light intensity in localized regions. The imperfections of a typical inclusion probably will lead to small increases in intensity. This may facilitate the onset of self-focusing when one is at a power density close to causing this effect.

Strains and Flaws

Almost any nonmetallic inclusion will differ in thermal expansion from the matrix sufficiently to generate large strains on cooling. Should the inclusion or its interface with the glass be weak, flaws and cracks are generated. This is definitely a weakening effect. Such effects are more severe with larger inclusions because there is a more abrupt property change in stress conditions at the interface and because there is a larger probability of finding a flaw. If thermal stresses generated by the pumping process approach the breaking strength of the glass, the additional stress or flaw from the inclusion could cause fracture. This effect apparently is not often observed. The next question is the influence of the laser light. Here, as stated above, some other mechanism must act in order to extract energy from the light beam. Therefore, strains and flaws become a secondary cause of damage and, hence, of secondary importance. The existence of fatigue effects in damage is a strong indication of the influence of flaws.

Electrostrictive Effects

In the presence of an intense electric field, additional photoelastic strain will occur around the impurity. The electrostrictive stress has a magnitude

$$p(de/dp)\,EE.$$

For a 100 joule/cm^2 pulse in 3 x 10^{-8} second, this stress amounts to about two atmospheres. The differential expansion around an inclusion less than 10^{-3} cm diameter should be negligible.

Plasma Generation

The mechanism for plasma generation in a nonabsorbing impurity should be very similar to that in the bulk glass. The mechanism of plasma breakdown is discussed more fully in the section on fundamental-damage mechanisms. The inclusion does not lower the threshold for damage significantly through this mechanism.

Linear Absorption Effects

Linear, or conventional, absorption effects will be important if the inclusion contains an impurity that absorbs at the laser wavelength. With the impurity at high concentrations within the inclusion, highly localized energy deposition may occur. This may generate the necessary breaking stresses. For small inclusions, with considerable solution at their boundary, small residual stresses may be expected. The temperature rise is given by (see Section VIII, Inclusion-Induced Damage),

$$\Delta T = (\phi\,\alpha) / c_v$$

where ϕ is the flux density, α the optical absorption coefficient, and c_v the specific heat. For a flux density of 30 joules/cm^2 and typical oxide specific heats, absorption coefficients greater than 10^2-10^3cm^{-1} are necessary to cause damage. This rules out many nonmetallic inclusions but is readily obtainable among compounds of the transition metals, among others. We conclude that a likely cause of damage by nonmetallic inclusions is linear absorption by highly absorbing inclusions. But, in most cases, nonmetallic inclusions will not cause damage.

Whatever the origin and nature of the inclusions that cause damage, the proper melting of optical-quality glass requires a generous injection of empiricism. A great deal can be learned from scientific approaches but these are too limited to provide a complete solution to the problems that arise. Experience in glassmaking and controlled experiments are necessary to supplement scientific and technological knowledge. In order that such empirical work be broadly applicable, it is helpful to have the new experience accumulate on one composition to reduce the number of variables as much as possible. During melting experimentation, the composition may have to be changed to achieve results but every effort should be made to force a composition decision as early as possible.

Conclusions and Recommendations

All approximate calculations indicate that metallic inclusions are more dangerous than nonmetallic inclusions. The only nonmetallic inclusions that appear dangerous are a highly selective class of high absorbers. Eliminating glass contact with metal through an all-ceramic melter seems more likely, after development, to consistently give laser glass that does not damage at low thresholds.

While the probability of eliminating damaging inclusions seems greater for ceramic melters, there remains the finite possibility that some ceramic refractories themselves may introduce detrimental inclusions. These may arise from high absorbtivity regions that dissolve out of the ceramic and are swept into the melt or from easily reducible metallic elements in the refractory.

It is recommended that a fundamental study be made of refractories and their propensity to generate inclusions in laser glass. A 2 man-year effort should establish significant results in this field. Capital requirements are nominal. Estimated cost, $150, 000.

It is recommended that present small-scale ceramic facilities be expanded and improved upon as the need for more high-quality glass arises and when the results of the above fundamental study warrant it.

VIII. INCLUSION-INDUCED DAMAGE

Introduction

Glass-laser rods often fail by the formation of interior cracks that run more or less normal to the direction of the beam. This type of damage usually occurs at energy thresholds much below those required for surface or tracking damage and, until quite recently, at least, it was the principal limiting factor on the performance of United States-made-glass lasers.

This internal cracking damage is thought to be initiated at solid inclusions in the glass. Direct evidence for this idea is not extensive but there are cases documented where the glass around inclusions apparently had melted in the laser beam and other cases where metal was found at the center of failure cracks. In addition, some correlation between the tendency to fail at low energy and the presence of metallic inclusions in the glass has been noted. The energy threshold for internal damage sometimes scatters considerably among specimens from the same stock and is higher in separated pieces of a failed specimen than in the whole specimen initially. These results indicate that there are internal sites, which are widely distributed in their tendency to initiate damage, and that the number density of the most damaging sites is quite small.

Mechanism

The general mechanism of damage was outlined in a paper by Bliss(13) and developed in detailed presentations to the panel by Gardon and Uhlmann. Uhlmann's presentation was based on a comprehensive paper on the subject by R. W. Hopper and D. R. Uhlmann(14) that has been submitted to the Journal of Applied Physics. Essentially, an inclusion with a high-absorption coefficient absorbs energy from the beam and becomes heated relative to the glass. The resulting tendency toward thermal dilation stresses the surrounding glass, sometimes sufficiently for fracture to occur. In fact, the pressure developed by the heating may induce a stress larger than the theoretical breaking stress, τ_{th}, of the glass.

The properties of the inclusion, which determine its tendency to cause this type of damage, are (1) absorption coefficient for the incident radiation, (2) emissivity, (3) physical dimensions and orientation to the incident beam, (4) thermal conductivity and heat capacity, (5) equation of state. The properties of the surrounding glass that will affect damage resistance are thermal conductivity and heat capacity and the distribution and nature of pre-existing microcracks.

It is expected that metallic inclusions are likely to be the most damaging type because they would absorb most of the incident radiation from a neodymium laser with negligible radiative loss during the period of irradiation. In the simplest limiting case, the particle absorbs energy at a rate proportional to its projected area on the beam (this ignores the correction for the Mie effect, which is discussed later) and, owing to the very high-metallic-thermal conductivity, it may be assumed for a certain range of particle sizes that (a) the temperature is roughly uniform through the volume of the particle at the end of the radiation pulse, and (b) there is negligible loss of heat by conduction to the surrounding glass during the pulse. It follows that the maximum average temperature, $\bar{T}_{max}$, of a spherical particle, taking its heat capacity to be constant with temperature, will be proportional to jt/r where

r = radius of the particle in microns,

j = power/unit area in gigawatts/cm^2

t = duration of pulse in nanoseconds.

For platinum particles, Hopper and Uhlmann$^{(14)}$ calculate:

$$\bar{T}_{max} \sim 800 \text{ jt/r in } {}^{o}K. \qquad (1)$$

This means that in a 30-nanosecond pulse of 20 joules/cm^2 the temperature of a particle 1 micron in radius would, ignoring absorption of energy in phase changes, reach 16,000^{o}K. The absorption of the heat of fusion would reduce this temperature only by about 600^o. It is seen that temperatures well above the normal boiling point of the metal might easily be reached. However, there is no mechanism for vaporization unless fracture is initiated in the surrounding glass. If the ratio of the thermal expansion to heat capacity is constant, the potential dilation ($\Delta V/V$) would be proportional to $\bar{T}_{max}$. The resulting tensile stress, τ_{max}, in the glass at the surface of the particle also would be proportional to $\Delta V/V$ and hence to $\bar{T}_{max}$. For typical glasses, Hopper and Uhlmann$^{(14)}$ estimate that $\tau_{max} \sim 60\ \bar{T}$ psi at the surface of a platinum particle. The theoretical fracture stress, τ_{th}, of the glass is expected to be of the order of 10^6 psi and, according to equation (1), this would be reached at the surface of a particle heated to 16,000^{o}K. Thus, for example, fracture would be initiated at the surface of any spherical-platinum particles with radii less than 1 micron, when exposed to a 30-nanosecond pulse of 20 joules/cm^2. We note that these calculations are based on extrapolation of the equation of state and other thermal properties of fluid platinum to temperatures far above any for which measurements are available. Existing correlations offer rather little guidance for these extrapolations. However, the principle of corresponding states for nonmetallic fluids predicts pressures at $\bar{T}_{max}$ at least as high as those calculated by Gardon and Hopper and Uhlmann.$^{(14)}$

In the limiting case considered, the maximum tensile stress at the particle surface climbs as the reciprocal of the particle radius. However, as the radius becomes very small, some effects become important that would limit and then reverse this trend. One effect is the sharp dropping off in the absorption efficiency when the particle thickness falls below the extinction length (~ 0.01 micron for platinum). Taking account of this and of the Mie effect(15) the ratio A_a/A_g of the absorption to geometrical cross-section of the particle, deviates significantly when a/λ, the ratio of the particle size to the wavelength of incident radiation, becomes quite small. Schalen's calculation of the dependence of A_a/A_g on a/λ for iron spheres is presented in Born and Wolf;(15) it indicates that A_a/A_g approaches 0 as a/λ approaches 0 but ranges between 1/2 and 2 for all values of a/λ greater than 0.2.

An effect, which may be more important in limiting the damage of small metal particles, is the increasing proportion of the energy lost by the particles to the surrounding glass as the particle diameter falls to values of the order of the thickness of the heat-diffusion zone in the surrounding glass. This thickness will be of the order of $(D_g t)^{\frac{1}{2}}$ where D_g is the thermal diffusivity of the glass and t is the duration of the pulse. For a 30-nanosecond pulse, $(D_g t)^{\frac{1}{2}}$ is of the order of 1,000 A. Also the glass layer around the particle may become too hot to support the tensile strength due to dilation. In this case the maximum stress will be developed at the boundary between the molten and solid glass. It will be due to the combined potential dilation of the metal and melted glass and will be substantially less than the maximum stress calculated on the assumption that the whole of the absorbed energy goes to heating the metal particle.

These considerations suggest that the most damaging platinum inclusions, from the point of view of initiating fracture, will be those in the radius range around 0.2 microns. It appears that particles with radii smaller than 200 Å would not be likely to initiate damage excepting at energy levels of the order of 100 or more joules/cm^{2}.

We now consider the problem of how far a crack initiated at the surface of the inclusion will grow. The stress should fall off with distance, d, from the particle surface according to the relation

$$\tau(r+d)/\tau(r) = (r/r+d)^3. \qquad (2)$$

The stress required to keep the crack running is $\sim\tau_{th}/2\ a(r+d)$ where a is the radius of the curvature at the root of the crack, usually assumed to be of the order of an atomic spacing. Therefore, the crack will run if

$$\tau(r)\,(r/r+d)^3 > \tau_{th}/2\ (a/r+d)^{\frac{1}{2}} \qquad (3)$$

For inclusions with radii of the order of a micron, this means that the crack diameter will reach about 5 to 10 times the particle diameter and then stop growing. The initial crack may not be large enough to cause failure. However, it is likely that the crack will be enlarged by each successive pulse. The reason is that the hot metal probably would run into the crack so that a much greater cross-section of metal would be presented to the next pulse. Thus, it appears that the crack would grow with each successive pulse until the metal is thinner than the thickness $(D_g t)^{\frac{1}{2}}$ of the heated-glass layer.

According to the foregoing analysis, the energy threshold for inclusion damage would increase with r for $r \gtrsim 0.1$ to 0.2μ. However, this result is based on the approximation that the temperature is essentially uniform through the particle at the end of the pulse. Actually, this approximation can hold only for particles smaller than about ½ micron. Owing to the finite thermal conductivity of the metal, the larger particles will develop a hot skin on their sides exposed to the beam while their interior will remain relatively cool. The temperature in the hot skin, and the resulting pressure at the surface, should be as large as that calculated for the smaller particles.

From these considerations it seems reasonable to conclude that all metal particles with sizes greater than some minimum, e.g., 0.1 micron radius for platinum, are likely to lead to serious damage at low-energy thresholds.

Any nonmetallic inclusions, which heavily absorb radiation of wavelengths in the range of that of the laser beam, also may cause damage by the foregoing mechanisms. However, most nonmetallic inclusions are relatively transparent to the radiation of the neodymium-glass lasers. Inclusions containing high concentrations of transition-metal ions would heavily absorb this radiation. It appears that ferrous compounds are the impurities that are most likely to occur in these inclusions.

Conclusions

To reach conclusions we shall consider the following questions:

1. What more would we like to know about the nature and mechanisms of inclusion damage?
2. How can the presence of damaging inclusions be better forecast and detected?
3. How can the introduction of damaging inclusions into the glass be prevented?

Concerning the first question, the experimental evidence linking interior damage to specific types (characterized by size, shape, and composition) of inclusions seems very limited. Because of the small number of failure events and the difficulty of finding and characterizing small inclusions, obtaining large amounts of such information would be a highly tedious and costly undertaking. On the other hand, the existing information seems reasonably consistent and more or less in accord with our expectations. Therefore, intensive studies to further demonstrate that certain types of inclusions, e.g., metal particles, directly cause damage probably should not be assigned a high priority.

The physical principles that govern the inclusion damage seem to be clearly enough understood. Also the methods for calculating the damage thresholds seem fairly well developed. Perhaps the principal limitation on these calculations is the lack of information and reliable correlations on the equation of state and thermal conductivity of the inclusions, particularly fluid metals, at the extremely high temperatures and pressures which they may reach at the end of the pulse.

The conditions that lead to the formation of damaging inclusions are considered in other sections of this report. It would be useful to develop methods for detecting potential internal-damage sites. The detection problem is a very difficult one, especially since failure could be caused by a single inclusion that might have a diameter no larger than a micron. Holographic methods appear to offer much promise for the solution of this problem and their further development and application should be encouraged.

Accepting that metallic and transition-metal compound rich inclusions are the cause of internal damage, the major problem becomes that of selecting glass compositions and methods of processing which minimize the formation of these inclusions.

IX. LASER-SURFACE DAMAGE

Introduction

There appear to be two possible mechanisms by which glass surface damage arises from high-energy laser irradiation even though it is almost universally agreed that plasma formation is present for both. It seems clear that the state of the glass surface is of paramount importance in generating and maintaining the plasma and that thermal heating by absorption of light at the surface is the controlling factor in plasma initiation. For example, it is observed that coatings of various kinds, which invariably enhance the effective surface-absorption coefficient, are unstable at high-radiation intensities and readily evaporate and contribute to the plasma. In addition, experiments indicate that immersion in liquids or exposure to different gases at various pressures have only marginal effects on surface damage. Such effects that are observed may be indirectly attributed to changes in the solid-surface characteristics by chemical interaction of the glass with the surrounding gas or liquid. Finally, a number of observations show that glass breakdown at the surfaces of internal bubbles is rarely, if ever, encountered. Over-all, the conclusion may be made that it is material from the glass surface that forms the plasma and that ambient gas or liquid plays only a secondary role in the damage process.

Plasma-Induced Mechanical Damage

One view of the mechanism for laser-surface damage has been proposed by Lubin(16) and is essentially mechanical in origin. The process may be described simply in that a near-surface plasma is formed so rapidly that local regions of the glass surface are subjected to explosive impact by shock waves from the expanding plasma. The local region initially undergoes Herzian compression with the buildup of tensile forces in the circumferential region surrounding the center of impact. If the tensile forces are sufficiently high, ring cracks may form. When the compression waves in the glass reflect and return as tensile waves to the original impact area, sections of this area may be ejected leaving a visible damage spot.

The experiments that led Lubin to this model are very instructive. A high-intensity laser beam was focused at an internal wall of an evacuated glass chamber, which contained electrodes that would sensitively measure electron densities arising from the ionization of neutral molecules. For the first few trials, it was observed that the damage threshold was relatively low and that the electron densities measured were sufficient to account for a plasma density, within a few millimeters of the irradiated surface, of 10^{17} ions/cm^3. This density would correspond to somewhat less than 1-millimeter-mercury pressure at standard-temperature pressure and was much higher than the evacuation pressure. It was concluded that molecular species left the glass surface and permitted the formation of the plasma. In addition, it was observed that the damage threshold continually rose with subsequent tests and this observation confirmed the concept that a source of easily vaporizable material was being cleaned, with each test, from the irradiated glass surfaces. The sequence of events for the over-all process would then seem to be thermal desorption, plasma formation with continued desorption, local impact, failure and ejection of glass particles.

Thermal Erosion

The second view of the damage process is that after initiation the plasma locally evaporates and erodes glass material by the bombardment of the surface with thermally excited ions. The process is visualized as being very similar to the "cathodic etching" process utilized by metallographers in preparing etched surfaces of refractory materials. There is one difference, however, in that it is highly likely that the material eroded and evaporated in the laser damage may significantly contribute to the energy absorption and subsequent growth and decay of the local plasma. Thus, it would be expected that the surface structure and composition, including absorbed chemical constituents, would play an important role in the erosion process.

Swain(17) has performed systematic studies of the effect of surface etching (chemical) on the damage threshold of laser glass. The intent of this work was to utilize etching to directly alter both the structure and the chemistry of the surface in a controlled fashion and to determine the effect of these changes on damage threshold. The most significant results from this work indicate that etching after resurfacing by polishing produces a much greater improvement in damage threshold than etching of an as-received glass surface. By and large, one can conceive of only one difference between samples in the original and resurfaced states and this difference is age. It is well known that glass surfaces react with and take up gases, particularly water and CO_2, and that these processes are diffusion-controlled and are continuous over long times. This effect may be termed aging and its presence is often indicated by troublesome outgassing of glass vacuum enclosures. It has been the subject of much study,(18) and in the present problem the results discussed in the previous section indicate it plays a role in laser damage. With respect to Swain's damage results, a qualitative but consistent explanation can be given in terms of thermal outgassing caused directly by laser radiation and/or by plasma heating.

Incomplete etching of an aged surface (i. e., etching to a depth that does not fully remove all the reaction products from aging) would leave material that could efficaciously contribute to plasma generation. One would anticipate that these materials would be the hydroxides and carbonates of alkali metals as well as hydrated silica and free water. It must be recognized that the surface one is concerned with in polished glass is neither plane nor defect free. It is one with innumerable flaws and cracks of variable depth filled with finely divided and highly reactive residue from the polishing operation. Continued etching of an aged surface would be expected to lead to improved damage thresholds until, at a point where only virgin glass is removed by the etch solution, a plateau in threshold would be achieved.

If etching of a freshly polished surface is performed, it would be expected that the degree of etching necessary to achieve a virgin glass surface would be much less than for the case of aged glass. In this case, the damage threshold should rise to the same plateau as described previously but in a much shorter etching time. The trend of the reported results seems to support this conclusion.

If the qualitative explanation of the particular results quoted above is substantially correct, then the thermal erosion model for damage contains many elements that are common to the mechanical damage model. The sequence of events would be thermal desorption, plasma initiation, plasma growth by high-temperature evaporation of glass components, and the continual formation of a damage pit by erosion and evaporation.

Conclusions and Recommendations

The present state of knowledge of the origin of surface damage in laser glass contains much speculation. There are, however, some very promising leads that demand further systematic experimentation. Further experiments, similar to those performed by Lubin with damage occurring on the inner surfaces of evacuated enclosures, could be incorporated in a program that would yield valuable information.

1. Test samples of varying glass compositions (fused silica, borosilicates with varying silica content, high-alumina glasses, various alkali metal-alkaline earth-silicate glasses) and of varying states of surface hydration (from steam-treated to superbly outgassed specimens) should be

used. In addition, it would be useful if the evacuation-test chamber be an integral part of a mass spectrometer so that the product species may be identified. We note that molecular-species identification would be one way of differentiating between the two models that are available (i.e., does the damage product contain gaseous Na_2O or NaOH molecules or silica monomers?).

2. The above procedure would be useful to perform critical experiments to determine whether or not thermal desorption of glass surfaces is a necessary condition for a low-damage threshold. A typical experiment would consist of thoroughly baking out and outgassing of a freshly prepared sample, then introducing one atmosphere pressure of inert and dry gas (argon) prior to the test. If the damage threshold were high, then desorption would seem to be an important factor. If the damage threshold were still low, then both models that have been proposed are somewhat suspect.

3. Scanning electron microscopy and microprobe analysis of surface-damage sites is mandatory. The morphology of these sites, as revealed in this fashion, should provide clear-cut evidence for model differentiation.

4. If mechanical breakdown is an important factor, then pre-stressing of glass surfaces by ion exchange or by thermal treatments may improve the surface-damage threshold. Laboratory experiments on hardened surfaces should be included in investigations of the effects of surface state on damage level.

The above kinds of tests to validate surface-damage mechanisms seem to be the first order of importance. It would also seem desirable to instigate a program for the selection of glasses for laser use which have been designed for high corrosion or weathering resistance. Such glasses would inherently exhibit good mechanical properties and would, in general, be more thermally stable. All these factors would be advantageous from the surface-damage standpoint. It is recognized that the attainment of these characteristics, in glasses ,which must exhibit many other demanding characteristics, is difficult. However, the exercise may be worthwhile for some compromises could be made.

In summary, the above recommendations propose two broad areas of work. It is felt that significant advances in surface-damage threshold cannot be made until the mechanisms of the damage process are fully understood. This is not presently the case and correction of this deficiency is required. Simultaneously, it would seem advisable to accept the preliminary findings that the chemical state of glass surfaces, altered by the environment, is an important factor in damage. Other glass systems or laser-component designs should be sought with the view to either achieving more stable glasses or for arriving at procedures whereby a given state of chemical stability may be maintained.

X. INTRINSIC BULK DAMAGE

Introduction

The nonlinear optical properties of solids, liquids, and gases have been studied extensively during the past decade. Numerous nonlinear processes have been identified and it seems clear that the processes induced by high-intensity light beams in glasses are not fundamentally different from those in crystals and liquids. An excellent introduction to the vast literature may again be found in papers presented at the proceedings of the symposium on damage in laser glass(19) and in a recent review article.(20)

In this section an attempt will be made to identify the most important mechanisms that constitute the fundamental limits for bulk damage on Q-switched glass lasers. Oral presentations(16) before the Committee on laser-induced plasmas, damage in alkali-halide crystals, and self-focusing mechanisms were very helpful for the following evaluation.

High-Intensity Electric Breakdown

It is well established that any material will disintegrate if subjected to a light beam of sufficient intensity for a sufficient duration of time. The final stage of this disintegration is the formation of a dense plasma. This plasma may be heated to thermonuclear temperatures, before it has time to expand, in an intense light pulse of short duration. The production of neutrons from pellets of LiD and solid D_2 evaporated by intense light pulses from Nd-glass laser systems has been reported.

The fundamental mechanism responsible for damage in an initially pure, nonabsorbing material is probably avalanche breakdown and/or electronic heating.(21) It starts from an initial conduction electron in the solid that is accelerated by the interplay of the collisions with optical phonons or other solid-state excitations and the electric field of the electromagnetic wave. It is the solid state analogue of the discharge mechanism in gases and liquids where the collisions occur with other ions. When the field is sufficiently intense, the electron may be accelerated so that it can create a new conduction electron by ionization or by creation of an electron-hole pair. The theory of gas-discharge formation by light waves, which is an extension of the formation of microwave discharges, is well developed and the process is known as the "inverse bremsstrahlen" effect.(21)

In a similar manner, Frohlich's theory for electric breakdown in crystals has been extended to visible frequencies.(22) The electrons impart a fraction of their energy to optical phonons, which, in turn, produce a heating of the lattice. Experiments have been performed in simple crystal structures such as the alkali halides. The observed threshold for damage in pure LiF for ruby-laser light lies at 5 x 10^{10} watts/cm^2, corresponding to 500 joules/cm^2 in a 10-nanosecond pulse. The theory predicts a threshold of 10^4 joules/cm^2 for this pulse duration. Similar numbers and discrepancies between theory and experiment occur for other alkali-halide crystals. The mechanism in glasses should follow the same physical principles and the threshold for avalanche breakdown should lie above 10^3 joules/cm^2 for a 10-nanosecond pulse. This appears to be in agreement with the observed energy density occurring in fossil-damage tracks.

Whereas in pure gases the first conduction electron to start the avalanche process has to be formed by multiphoton ionization, in a crystalline solid or a glass it is most likely that a small number of carriers is always present due to thermal or simple photo-ionization of some shallow traps.

Several other possible damage mechanisms in a pure glass-host material, in the absence of inclusions, are conceivable such as heating by multiphoton absorption and stimulated Brillouin scattering. The damage threshold for these processes appears to be higher and the experimental evidence also suggests that, for the pulse durations and laser glasses of interest, they do not set a fundamental limit by themselves although they may occur simultaneously with the above-described breakdown mechanism. This mechanism usually leaves a fossil track of resolidified material. The shock wave generated by the sudden release of energy in a confined volume often produces pitting or cratering at the output face of the laser rod. This material damage occurs when the shock wave reaches the output face after the laser pulse has passed. Incandescent radiation from the heated regions has also been observed. The intrinsic-threshold damage is sufficiently high that existing laser pulses have to be focused to achieve the threshold intensity. The threshold for bulk electric heating and breakdown by itself does not impose a serious limitation for the operation of Q-switched glass lasers.

Self-Focusing

A most important observation is that focusing may take place in an unavoidable manner inside the laser rod. This phenomenon of self-focusing is well established and was, in fact, first observed in glasses by Hercher in 1964. The threshold for its onset has been measured in many different materials for several values of pulse duration, beam diameter, wave length, and temperature. For pulses longer than 10^{-9} second, the dominant mechanism for self-focusing in glasses is most likely the electrostrictive effect, which may also be considered as a stimulated near-forward Brillouin scattering, in which the acoustic wave travels transverse to the direction of laser propagation. The effect can fully develop in a time equal to the diameter of the laser beam divided by the acoustic velocity. For shorter pulse duration, the self-focusing action will be weaker and have a transient or incipient character. For laser pulses of picosecond duration, the self-focusing may take place through a quadratic Kerr effect or through a combination of absorptive heating and the intrinsic-temperature dependence of the refractive index $(\partial n/ \partial T/_{\rho} > 0)$. In laser glasses the former effect is probably dominant. The quadratic Kerr constant for fused quartz has been experimentally determined to be about 10^{-13} esu (electrostatic units). The corresponding index change is $10^{-13}\,(E)^2$. For the intensity gradients $\Delta(E)^2$ prevailing in picosecond pulses, this could indeed account for the observed self-focusing and filament formation.(23) The quadratic Kerr constant may, besides a contribution from purely electronic origin, also in part be determined by librations of molecular groups. There is not sufficient data in a variety of glasses to separate these mechanisms. The answer to this question would be of importance for the operation of picosecond-pulse, glass-laser amplifiers.

Once the light intensity has been sufficiently concentrated by the self-focusing mechanism, the fundamental-damage mechanism, described in the preceding paragraph, takes over resulting in fossil-damage tracks of a filamentary nature. If an absorbing inclusion happens to lie in the self-focused region, the damage will be more extensive.

Quantitative results with diffraction-limited, laser pulses of 50-nanosecond duration in dense-flint glass show that the energy threshold for self-focusing may be as low as 10 joules/cm^2, corresponding to 2×10^8 watts/cm^2. These results were, however, obtained with a laser beam with a diameter smaller than 1 mm, but they certainly indicate that a self-focusing in the bulk laser-glass material poses the most serious fundamental limitation, by its very nature, very sensitive to the spatial and temporal structure of the laser pulse. What usually happens is that a fraction of total energy gets focused in a filament, while most of the laser pulse passes through unperturbed. The fossil filamentary-damage track vignettes only a small part of the next pulse and the laser rod may be reused for many subsequent shots. It is interesting to note that for beams which have a homogeneous spatial intensity profile, the filaments form preferentially at the periphery of the beam where the transverse light intensity gradient is largest.

For damage to occur, the self-focusing has to proceed to such a degree that the breakdown limit is exceeded. The fundamental limitation of the bulk laser-glass material lies, therefore, in the development of self-focused regions.

The details of self-focusing depend on the bulk properties such as the elastic moduli and the index of refraction, which determine the electrostrictive effect. These parameters cannot be varied over wide ranges but a factor of 2 or so improvement in the threshold for self-focusing may be obtainable. An effort to develop a laser glass with optimum properties to raise the self-focusing limit appears warranted, since the threshold for filamentary-track damage has already been reached in a few selected configurations.

It is expected that Er^{3+}-doped, glass-laser materials, operating at longer wavelengths will show similar damage behavior. They may present additional problems due to absorption from different rare-earth ions in higher concentrations.

It should be kept in mind that the formation of a self-focused region within the rod depends not only on the intensity but also on the dimensions and transverse-mode structure of the beam, on the pulse duration and temporal structure within the pulse, and on the length of the rod. No experimental damage results are available for homogeneous beams of large diameter and pulse durations of 10 to 30 nanoseconds. Since the acoustic wave can travel only a fraction of a millimeter in this duration, a transient analysis of the incipient formation of the self-focusing appears to be in order. Since the initial nonlinear-index variation, responsible for the onset of self-focusing, is weak in beams with a nearly flat intensity profile, the theory for the ideal linear medium would have to be modified to take account of the inevitable small spatial fluctuations in the linear index of refraction.

Further experimental data for incipient self-focusing characteristics in Q-switched beams with larger cross-sections and for the dependence on geometrical parameters of beam and rod are needed. An educated guess based on the presently available theoretical and experimental information is that the threshold for the formation of a self-focused region in the geometry prevalent in existing Q-switched, glass-laser devices lies between 50 and 100 joules/cm^2.

Self-focusing may well become a bottleneck at the flux density desired in laser operation but its effects could be mitigated by geometric design incorporating shorter rods or slabs.

Conclusions and Recommendations

1. A fundamental limit for damage from Q-switched pulses in ideal bulk glass-laser material is set by the self-focusing through the electrostrictive effect followed by avalanche-electric breakdown and electronic heating in the focused region.

2. The threshold for self-focusing in Q-switched, glass-laser systems tested to date lies between 50 and 100 joules/cm^2 for 30-nanosecond pulses. It does not vary widely with composition. The threshold does depend, however, sensitively on the beam size, geometrical and temporal structure of the pulse, and on the geometry of the laser rods or slabs. The threshold for avalanche breakdown exceeds 1,000 joules/cm^2 for pulses longer than a nanosecond.

3. It is recommended that further theoretical and experimental studies be made of the self-focusing and mode distortion as a function of temporal and spatial structure of the beam and the geometry of the output elements of the Q-switched laser. The study of self-focusing and damage in the picosecond-pulse regime should also be continued. An effort of 4 man-years is recommended. A minimal healthy effort would include 2 man-years.

4. Raising the fundamental threshold for self-focusing and damage in the bulk material by changing the material properties of the bulk glass, such as elastic moduli and optical index, should be pursued with lower priority because the maximum factor for improvement by extensive and expensive glass-materials engineering is not expected to exceed a factor of 2.

XI. FACILITIES AND DIAGNOSTICS FOR TESTING DAMAGE IN LASER GLASS

Introduction

In any effort to improve the damage resistance of laser glass, it is essential to develop adequate test facilities and suitable diagnostic procedures to ensure that the dominant mechanism of damage is identified, and that reliable and reproducible testing procedures can be developed. In preparing this report, information was gathered from three sources. The first was the ASTM Symposium on Damage in Laser Glass held in Boulder, Colorado in 1969.(24) The second source was testimony taken in the sessions of the National Materials Advisory Board Ad Hoc Committee on the Fundamentals of Damage in Laser Glass, and the third was the written response to a survey mailed out to the principal laboratories involved in the development and testing of high-powered glass lasers and damage-resistant glass.

Existing Facilities

At present, there are two manufacturers of high-damage threshold laser glass in the United States, The American Optical Corporation in Southbridge, Massachusetts, and Owens-Illinois, Inc., in Toledo, Ohio. Each of these companies has started the development of damage testing facilities. At American Optical, two test devices have been constructed. A high-brightness device, Q-switched with a Pockels cell, radiates about 90 joules in a 10- to 30-nanosecond pulse, with 0.04-milliradians (mrad) beam divergence from a 7.1 cm^2 aperture. Due to the excellent beam quality, the output of this device can be doubled in frequency with high efficiency, yielding 23 joules at 0.53 μm, and 2.7 joules in 0.27 μm. An additional amplifier, approximately 30 cm^2 area, gives an output of 300 joules in 0.04 mrad. An amplified spontaneous-emission generator has been constructed expressly for quality control in the testing and fabrication of laser rods. This device emits 80 joules in a pulse of about 10-μsec duration from a 7.1-cm^2 aperture into a 1-mrad beam. The pulse width of this device can be varied continuously from 100 nanoseconds to 10 microseconds. This device is described in detail in Reference 24. At Owens-Illinois, Inc., a Q-switched, Brewster angle oscillator-amplifier system has been constructed with a 3-cm^2 beam cross-section. This system emits 120 joules in a beam of 2-cm^2 outside cross-section in 55 nanoseconds with a beam divergence of 1.0 mrad times 2.0 mrad (elliptical beam). These test facilities were developed by the glass manufacturers for the express purpose of quality control and materials specification.

The Compagnie Generale d'Electricite (CGE) in Marcoussis, France, has concentrated an extensive effort on the development of high-brightness glass lasers in the last five years. These devices are now available in the United States through Hadron, Inc., of Westbury, New York. The presently existing VD/VK 640 system radiates a maximum of either 250 joules over 32.6-cm^2 aperture in 5 nanoseconds in a 3 x 5 mrad beam or 500 joules over 32.6-cm^2 aperture in 30 nanoseconds in a 1 x 2 mrad beam.

In response to the survey questionnaire, representatives of American Optical, Owens-Illinois, Inc., and Hadron indicated above have indicated a willingness to entertain proposals for the use of their facilities for damage testing under contractual agreement.

There are two government laboratories with current programs in testing damage in laser glass, the National Bureau of Standards in Gaithersburg, Maryland, and the Naval Research Laboratory in Washington, D. C. The National Bureau of Standards has installed a Brewster angle neodymium-glass system consisting of an oscillator followed by two amplifiers, with a 3 cm^2 in side beam area. It radiates 30 joules in 30 nanoseconds in a beam of 2 cm^2 outside cross-section in 30 nanoseconds. This glass system is not presently in operation but is undergoing acceptance tests. The Naval Research Laboratory (NRL) laser system is presently a CGE VD 320 system, capable of radiating 117 joules over 7.2 cm^2 in 30 nanoseconds in a 0.2 mrad. It is used both for plasma-production experiments and for glass-damage testing. Work is underway to extend the capabilities of this laser to higher energy.

In any of these systems, higher-energy densities than those quoted can be achieved either by focusing the output beam into the sample (passive test) or by recollimating the beam at a smaller diameter than that indicated with an afocal telescope. In the case of the CGE systems, for example, afocal-beam expanders are normally placed between successive stages of amplification. By removing the last of these, an energy density as high as 25 joules/cm^2 can be achieved in the VD 320.

The Air Force Cambridge Research Laboratory (AFCRL) has an active program in damage testing in ruby lasers with special emphasis on short-pulse phenomena. The ruby-laser system at AFCRL will, when completed, radiate 1 to 10 joules over a 1.27-cm^2 aperture in a time from 0.1 to 2 nanoseconds. The system will also be capable of normal Q-switched operation at higher energy.

Although no systematic investigation of laser damage is underway at the Air Force Weapons Laboratory (AFWL) in Albuquerque, New Mexico, an extensive program in high-power laser development is being carried out there. Damage tests have been run at 20 joules/cm^2, using a device radiating 60 joules over 3 cm^2 in 40 nanoseconds. The AFWL is planning to extend their capability to the level of a few hundred joules in a pulse of variable duration, from a few nanoseconds to 30 nanoseconds.

Four laboratories, whose primary emphasis is on the application of high-intensity glass lasers, have operating devices that could be employed for occasional damage tests. These are the Lawrence Radiation Laboratory (LRL) in Livermore, California, United Aircraft Research Laboratory in Hartford, Connecticut, University of Rochester, in Rochester, New York, and the Sandia Laboratory in Albuquerque, New Mexico. The performance characteristics of the lasers extant or planned at these laboratories are summarized, along with the specifications of the devices described above, in the summary table.

In most of the devices described above, the sample can be substituted for one of the amplifier rods in the system for active testing (sample pumped). For testing an umpumped sample, the sample can be placed outside the laser system with the beam either unfocused, recollimated, or focused into the sample.

In summary with regard to facilities, each of the three principal manufacturers of laser glass is developing facilities for testing glass rods as active elements of a well-controlled amplifier system. One government laboratory, NRL, has an ongoing program in damage testing in a high-brightness device but this device is only available for damage testing on a part-time basis. The National Bureau of Standards (NBS) is developing a comparable testing capability but has not completed device installation. Sporadic damage observations are being carried out in other government laboratories but no systematic program of damage testing exists other than at NRL and NBS. The program at AFCRL addresses itself to a problem of damage in the sub-nanosecond time regime, and is carried out at the ruby wavelength. No high-energy damage test facilities exist at present for Erbium laser glass.

Passive Properties of Glass

In obtaining information on the mechanism of damage and for the development and design of laser systems, it is essential that a full description of the glass properties be obtained. General optical properties, mechanical, and thermal properties such as index of refraction, absorption, scattering, and birefringence, heat capacity, thermal conductivity, and elastic moduli, should be well described both before and after samples are tested. The chemical composition of the glass sample is significant as well, since impurities such as iron can contribute significantly to the observed absorption. The ionic state of the impurity (FE^{2+} or Fe^{3+}) is significant in determining its role in the various possible damage mechanisms.

The National Bureau of Standards has the capability of providing these data in a uniform and systematic way. An effort of 8 man-months was made to measurement of bulk-physical properties of glass in 1969 out of a total of about 17 man-months spent on laser damage as a whole. A slightly larger effort is anticipated in 1970. A report has been issued(25) by NBS on tests run on several samples of laser glass.

Pulse Duration and Beam Profile

There are three basic areas of investigation in regard to laser damage: damage due to microinhomogeneities (including inclusions), surface damage, and damage due to beam trapping. These different damage phenomena are associated with different physical mechanisms and are each characterized by different time regimes. It is thus possible to separate the effects of these several phenomena by varying the duration of the experiment. As an example, the amplified spontaneous-emission generator constructed at the American Optical Company is designed explicitly to operate in a long-time regime so as to eliminate beam trapping and thereby test the presence of particulate inclusions and inhomogeneities. Those devices listed in the table which operate in the sub-nanosecond regime, on the other hand, will be sensitive to beam-trapping damage rather than inclusion-induced damage.

The onset of self-focusing is strongly dependent on the presence of gradients in the radial-intensity profile of the beam. For this reason, it is essential that the radial-intensity distribution be known for each test device, along with the temporal variation of the intensity. In sub-nanosecond pulses, or, in fact, in any arrangement where electro-optic shutters are employed, a substantial fraction of the laser energy may be contained in a part of the pulse transmitted at low-power levels. Such "leakage" is recorded in a calorimetric measurement of pulse energy, but not in a pulse-width measurement, so that the computed average power for such a pulse may be grossly over-estimated.

Sample Preparation

For tests conducted external to the laser cavity, such as either passive tests of bulk properties, or unpumped-damage tests, there is considerable variation in size and preparation of samples. It is essential, if tests are to be reproducible, that great care be taken in specifying the experimental arrangement (angle of incidence, plane of polarization, focal geometry) and the sample condition (surface preparation, coatings, previous exposure of sample). In the work reported before the Committee on surface damage, for example, an order of magnitude change in damage threshold was noted on variation of the surface condition(26) and on repeated exposure at power densities below the damage threshold. (15)

Where the sample is to be pumped as an integral part of the laser system, it is general practice to substitute the test specimen in rod form in the system. It is suggested that the development of segmented-laser assemblies for test devices would allow the use of smaller samples with correspondingly lower fabrication costs. Thus, the sample to be tested could be substituted as a single element of a high-power segmented assembly. Difficulty in alignment and assembly of segmented devices may limit the practicability of this procedure. Testing of small samples is not applicable to the detection of particulate inclusions that may be dispersed in the glass in low concentrations.

Diagnostics

There are three levels of diagnostic observation relevant to damage testing: observations before testing, during irradiation, and after testing. The most commonly used diagnostic technique at present is the inspection of samples prior to testing, by large-angle light scattering, to reveal the presence of inhomogeneities of a size comparable to or larger than the wavelength of the incident light. Additionally, interferometric and Schlieren tests can reveal striae, strains, and other refractive index anomalies, and are in general use. Absorption spectra of samples are useful in describing the glass but generally cannot be run on full-sized laser rods.

The value of the absorption constant at the laser wavelength, which is extremely important in assessing the ultimate laser performance, is typically measured on a full-sized laser rod. In general, it is desirable to obtain as complete a description as is possible of the bulk properties of the glass being tested.

For surface damage, it is customary to make a careful inspection of the surface prior to testing to detect in advance any pits or flaws in the surface condition, especially if the surface is coated.

Perhaps the most specific information about the damage mechanism can be obtained by those observations made during the damage process. The most important diagnostic here is spectral emission from the damage site, particularly in the case of the formation of microplasmas. Acoustical energy has also been detected as a concomitant of inclusion-induced damage. If the mass and optical spectrum of the plasma formed in the process of surface damage could be observed, valuable information could be obtained regarding the nature of the damage process. Photoelectric emission from the surface has been measured both at CGE and the University of Rochester, and, in general, there is a good correlation between the observations of surface damage and a high value of photoelectric current from the glass surface.

Postmortem examination of laser-glass samples can also reveal substantial information about the diagnostic process. If it is suspected that inclusions of a particular material are localized at the damage sites, microprobe techniques could be used to confirm this suspicion. The geometric arrangement of damage tracks is also an important diagnostic. For example, in cases of damage incurred at picosecond-pulse lengths, the damage tracks generally take the form of lines of very fine bubbles, which are considered evidence of the role played by beam trapping in the damage mechanism.

There is an essential distinction between the identification of platinum inclusions in platinum-melted glass and that of identifying other microinhomogeneities. In the former case, the principal question is the size of the particle, while in the latter case the composition and nature of the inhomogeneities are in question. In the extremely homogeneous glass recently made, damaging inclusions are very small leading to great difficulties in postmortem analysis of damage sites. Some promise in localizing damage sites is afforded by the use of holographic examination.

The two principal domestic laser glass manufacturers have undertaken relatively ambitious and painstaking investigations of the damage process. At Owens-Illinois, Inc., a sample of glass has been subjected to microscopic examination prior to irradiation, and the location of all detectable particles and inclusions has been mapped out. The sample was then irradiated to induce damage and the location and nature of the damage sites were compared with the original map of the location of inhomogeneities. The data are currently being reduced but preliminary results indicate that a simple correlation may not exist. At American Optical, a holographic technique has been developed to detect small index changes during the period of sample irradiation. A film record is obtained of the holographic image at the exit face of a glass-rod sample. This technique should allow the observation of stress concentrations developed during and immediately after the damage process. The technique of real-time holography also has been suggested as a means of comparing sample condition before and during damage testing.

No catalogue of diagnostic techniques is really complete. As specific theories of damage mechanism develop, new diagnostic methods logically evolve from them. It is clear, however, that as complete a characterization of the sample under test as can be obtained is essential in interpretation of the test results. Equally important is a complete knowledge of the radiation characteristics of the test laser. The two industrial laboratories, AO and O-I, and the government laboratories, NRL and NBS, most heavily involved in developing test devices, have gone to great lengths to obtain a full characterization of the spatial distribution of energy in the test-laser beam. This is absolutely essential if the test results obtained are to be meaningful.

Conclusions

At present, each of the two principal domestic manufacturers of laser glass has undertaken the development of facilities for the test of damage in laser glass at high power. American Optical has developed a variable pulse-length device, concentrating on the detection of damage arising from particulate inclusions, as well as a device operating in the normal, Q-switched regime. The Owens-Illinois, Inc., device operates in the usual Q-switched regime so that either bulk or surface damage can be induced, depending on the experimental configuration.

Among government laboratories, only the National Bureau of Standards and the Naval Research Laboratory have undertaken any systematic investigation of damage in laser glass. The NBS program is more extensive in terms of man-hours expended but has been concentrated to date on developing passive tests for the characterization of laser glass. The NRL program, although part-time in nature, has progressed further in active testing due to the availability of a suitable test device.

There are several other devices in the United States that could be employed for damage testing purposes but no systematic program exists for laser glass damage studies outside the laboratories mentioned above. There are several laboratories where programs relevant to damage studies are underway. A study of the influence of surface treatment on surface-damage threshold has been carried out at the Lawrence Radiation Laboratory at Livermore, and a study of the influence of pre-irradiating the surface to raise the damage threshold has been carried out at the University of Rochester. The Air Force Weapons Laboratory is planning to support a study of the use of ion-implantation and electron-beam polishing to raise surface-damage threshold. These are a few examples of supporting studies which exist or are planned.

A full description, both of the laser-glass material and of the radiation pattern of the test device, is required if meaningful results are to be obtained from damage tests.

For testing damage thresholds in the picosecond-time regime, no facility presently exists solely devoted to this purpose. Each of the principal laboratories involved in the construction of a large, sub-nanosecond-pulse laser has done some damage testing, and will continue to do so, but no attempt has been made to standardize testing procedures. Extreme caution is needed in the calorimetry of sub-nanosecond pulses.

The cost of establishing adequate test facilities is considerable. To purchase or construct a device comparable to the VD 320, costs from $100,000 to $150,000 for the device alone. Additional expenses include a highly-skilled technician to keep the device in alignment and to set up various test configurations and, at the least, a half-time professional. Thus, to establish and run a minimal routine testing program costs in the order of $200,000. In the laboratories of the glass manufacturers, the cost of establishing a suitable test program can be included, in part, as a portion of the cost of manufacture. It is in this spirit that AO has developed its long-pulse test device, and O-I, its Q-switched test amplifier. To carry out exhaustive studies of damage mechanisms using these facilities will require further funding, some of which might be expected to come from government sources of support as it has in the past.

The role of test facilities in the government laboratories is somewhat different from that of the manufacturers. Government facilities must exist in order to provide reference standards for damage testing. Since the government laboratories can be viewed as disinterested parties by the manufacturers, impartial testing can be carried out in these laboratories. It is possible to specify acceptance standards in terms of standard tests run on government test devices. Additionally, a great deal of exploratory research on damage mechanisms can be carried out in government laboratories. At present, the NRL test device is the one device available in the government for testing and research of the kind described above, albeit on a part-time basis.

As part of a program for the development of laser standards, Subcommittee II of Committee F-I of the American Society for Testing and Materials has undertaken the task of developing damage standards and specifications. A Symposium on Laser Damage was held in Boulder, Colorado in June of 1969 and another is scheduled for June of 1970. In the ASTM Committee, a concentrated effort has been made to bring together representatives of the laser materials industry, laser manufacturers, and government users, in order to establish useful and realistic specifications.

Recommendations

It is recommended that existing programs in the development of test facilities, both at the O-I and AO laboratories and at the NBS and NRL, be continued. A close coordination should be encouraged between NRL and NBS laboratories with strengths in this area that are more complimentary than competitive. It is also strongly recommended that the development of test facilities, specifically addressed to the question of damage in laser glass, be closely coordinated with the ASTM activity.

Presently existing test facilities are adequate to support the development of glass exhibiting a higher-damage threshold for 30-nanosecond pulses. It is strongly recommended that increasing emphasis be placed on developing test facilities and procedures for sub-nanosecond damage studies. Since the total energy required for such studies is not as great as 30 nanoseconds, the capital investment required to set up a laser is considerably less, of the order of $20,000 to $50,000, depending on the configuration. It is recommended that the laboratories presently engaged in materials testing be encouraged and, where necessary, funded to modify existing oscillator-amplifier combinations to allow the testing of glass in the sub-nanosecond regime. Some funding should be made available to support fundamental investigations of beam trapping in glass, and a relief of beam trapping by appropriate engineering design.

A minimum of $250,000 a year should be set aside for the maintenance of test programs at NRL and NBS, supporting studies in other laboratories, purchase of materials, and modification of existing test devices. Close coordination must be maintained with device development in the AEC and with the ASTM standards effort.

TABLE II

OCATION	ENERGY (Joules) * Note 1	PULSE DURATION (nsec)	BEAM AREA (cm^2) * Note 2	DIVERGENCE FULL ANGLE (mrad)	PUMP ENERGY (Kilo Joules)	MODE OF SWITCHING	COMMENTS
merican Optical	90	10 - 30	7.1	.04	40	Pockels	Diff. Limited
	300	10 - 30	30	0.04	120	Pockels	Variable Pulse Length
	100	10^2-10^4	7.1	1.0	120	Kerr Cells	Amplified Spontaneous-Emission
vens-Illinois	120	55	2.0	1.0 - 2.0	35	Rotating Prism	
adron	250	5	32.6	3.0 - 5.0	206	Kerr Cell	
	500	30	32.6	1.0 - 2.0	206	Rotating Prism	PTM
BS	30	30	3.0	2.8	30	Pockels Cell	Undergoing Acceptance Test
RL	117	30	7.2	0.2	60	Rotating Prism	
FWL	60	40	3.0	1.5 - 3.0	40	Rotating Prism	
nited Aircraft	10	0.1 - .001	11.3	0.1	60	Dye Cell	Pulse Selected
RL	100	.5 - 200	5.0	0.2	200	Pockels	
	10	.01 - 0.1	5.0	1.0	100	Dye Cell	
ndia	180	30	10	1.0 - 2.0	80	Pockels	
	75	.003	10	1.0 - 2.0	100	Dye Cell	
niversity of ochester	250	0.15	20	-----	200	Dye Cell	Pulse Selected

ote 1: Energy measurement is uncertain in pulses shorter than 1 nsec.

ote 2: Beam Area in normal configuration. Area can be reduced for specific tests.

lank spaces indicate information not collected at time of preparation.

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13. ABSTRACT

In spite of the fact that substantial amounts of money have been expended by both industry and the military, optically induced damage in glasses still limits the usefulness of laser-glass devices. It is believed that the existing material limitations are not insurmountable. It is therefore desirable to evaluate and review the efforts in various industrial and governmental laboratories to define the fundamental material limitations and to indicate steps in research and development necessary to reach those fundamental limits.

The report of the Committee outlines the several problems that need to be overcome to permit production of a laser glass having a damage threshold of 100 joules per cm^2 while possessing the other qualities requisite for good high-durability laser glass. A program directed toward the solution of these problems is documented in the report.

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	ROLE	WT	ROLE	WT	ROLE	WT
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Composition						
Manufacturing						
Metallic Inclusions						
Nonmetallic Inclusions						
Surface Damage						
Bulk Damage						
Inclusion-Induced Damage						
Testing for Damage						