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Platinum Particles in the Nd:doped Disks of Phosphate Glass in the Nova Laser

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ABSTRACT

The disks of Nd:doped phosphate glass in the amplifiers of the Nova laser contain platinum particles with sizes ranging from < 5μm (detection limit) to about 100μm. The particle density varies from about 0.01 to 1.0 cm^{-3}. These particles cause fractures when irradiated at fluences > 2.5 J/cm^2 delivered in 1-ns, 1064-nm pulses. Under repeated irradiation at 5-7 J/cm^2, damage from small (< 5μm) particles asymptotically approaches a limiting size, but damage surrounding the larger particles grows steadily. The damage threshold fluence, 2.5 J/cm^2, corresponds to operation of Nova at one-half the desired output for pulse durations longer than 1 nsec. Operation at higher fluences causes accumulation of damage in the output amplifiers and requires replacement of the disks in those amplifiers on an accelerated schedule.

Large-area damage tests have been used to characterize samples from recent melts made to reduce the number of platinum particles. The 500-J, 1-ns pulses available at the midpoint of an arm were used to produce fluences of 7-10 J/cm^2 over test areas 6-9 cm in diameter. Similar irradiations of areas 25-30 cm in diameter were accomplished using 5-kJ, 1-ns pulses available at the output of an arm. Preliminary results from these experiments indicate that a significant reduction of the inclusion density can be obtained through changes in the vendor's glass melting conditions.

Key words: platinum inclusions, phosphate glass, borosilicate glass, large-area damage tests.

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I. Introduction

Isolated bulk damage sites caused by laser irradiation of platinum inclusions have been observed in the components of the Nova laser and its predecessor, Novette. These damages occurred in amplifier disks made of Nd-doped phosphate laser glass and in lenses made of borosilicate glass. This problem was first encountered in operation of Novette, a laser with two beam lines (each arm with 10 kJ, 1 ns output), each identical to one of the ten beams of Nova.

The output stage of a beam line arm of either of these lasers contains groups of Nd:glass amplifiers with apertures of 20.8, 31.5 or 46 cm (Fig. 1).[1,2] Each group of amplifiers is followed by a vacuum spatial filter. A laser pulse passing along the beam line is amplified at one beam diameter to the desired pulse energy, and then focussed into a spatial filter which removes small-scale spatial ripple and enlarges the beam diameter to properly match that of the next group of amplifiers. A spatial filter at the output of the 46 cm amplifiers provides a final cleaning of the beam and increases its diameter to 74 cm which is maintained over the final path to the target chamber.

As a result of this series of amplifications and beam expansions, the variation of pulse energy or beam fluence with position along the chain is an approximately sawtoothed function (Fig. 2), with the largest fluence occurring at the output of the last 46-cm amplifier. During generation of 10 kJ, 1-ns pulses, the average fluence over the effective area of the 46-cm beam (1330 cm^2) is 7.5 J/cm^2. Because this large fluence exists at the input to the final spatial filter, the fluence at the maxima of beam ripples may be more than 1.5 times the spatially

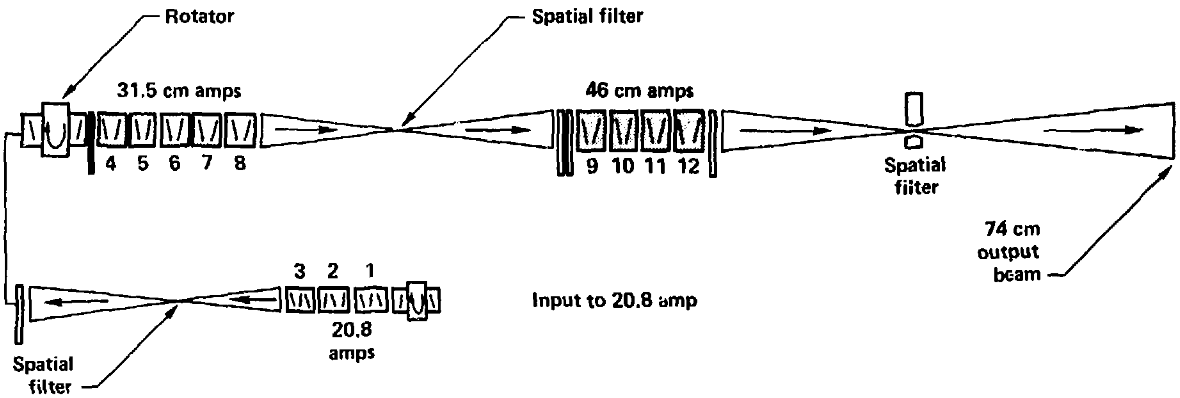

Fig. 1. Schematic diagram of the final 3 amplifier stages (20.8 cm, 31.5 cm and 46 cm) and spatial filters that are a part of each one of the 10 beam lines on Nova.

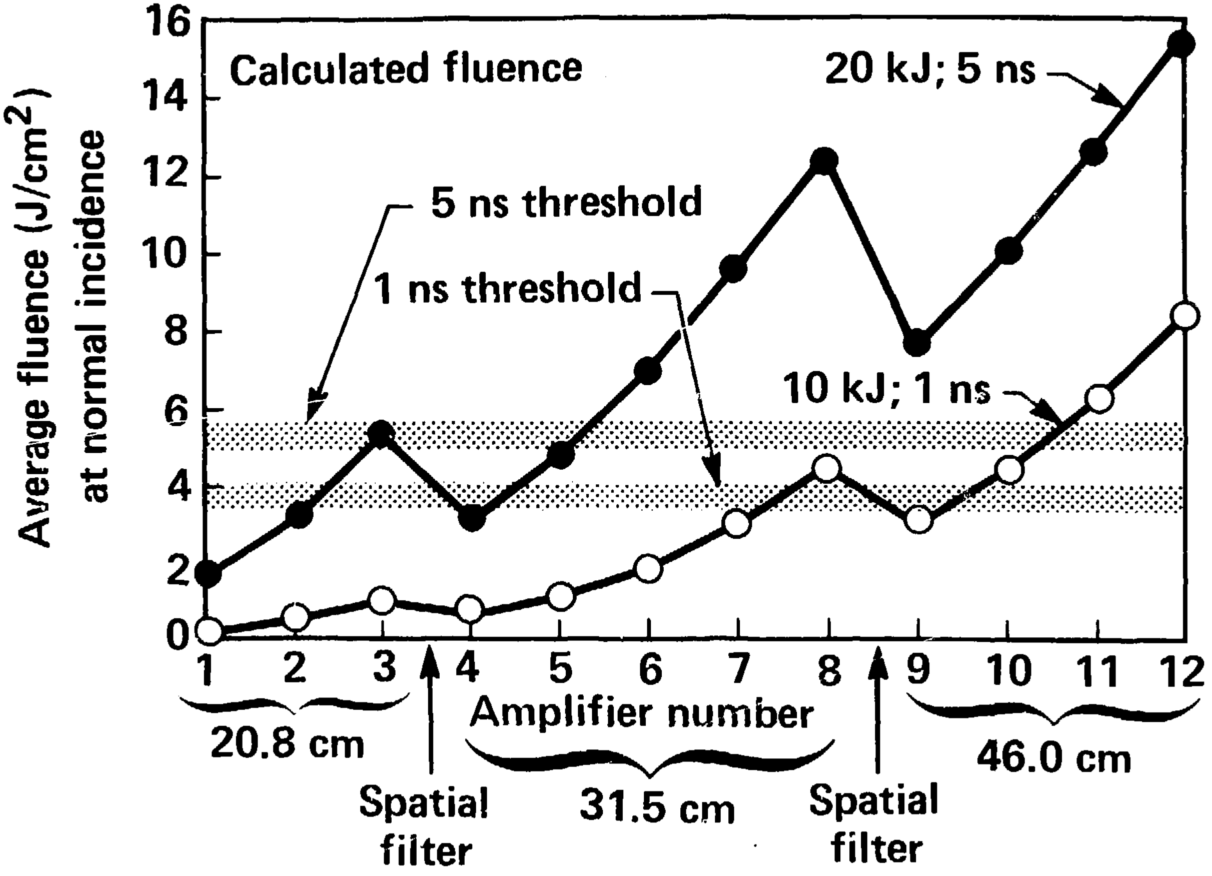

Fig. 2. Beam fluence at normal incidence as a function of position along the final 3 amplifier stages of a Nova beam line.

averaged fluence. The components in this region of high fluence are the output 46-cm amplifier disks and the 46-cm input lens to the final filter. The lens is positioned normal to the beam and experiences the full beam fluence. Note, however that the fluence is lower in the 46-cm amplifier disks because they are positioned at Brewsters angle.

Internal damage in a 46-cm spatial filter lens occurred during the initial full power pulses generated by Novette. Several small volumes each containing an internal damage were cored from the lens and broken to expose the centers of the damages. When these locations were examined by scanning electron microscopy, each was found to contain a platinum particle.

Because the glass was purported to be platinum free, this failure was initially believed to be an isolated incident caused by an identified degradation of a platinum stirrer used in production of the glass. However, replacement lenses made from glass purchased from two other vendors also damaged, indicating that platinum was probably present in most of the available homogeneous glass, all of which had been produced in platinum crucibles. The problem of damage to borosilicate glass was circumvented by fabricating the heavily loaded lenses from high purity fused silica. To date, bulk damage has not been detected in these silica lenses.

A more serious problem arose when bulk damage was observed in the phosphate glass amplifier disks in Novette. This problem could not be solved by substitution of materials because all presently available laser glasses are melted in platinum. A group of chemists, physicists and engineers was assembled to address the task of eliminating platinum.

There were five major goals of the group:

(1) measure the thresholds for damage to selected individual platinum inclusions and the rate of growth of the damaged volume surrounding the inclusions,

(2) develop an accurate model for the damage caused by laser heating of Pt inclusions,

(3) assess the damage on Nova and determine a safe operating range during the period of amplifier repair,

(4) develop a reliable quality assurance technique for inspecting large volumes of glass for the presence of Pt inclusions, and

(5) work with the glass vendors to quantify the mechanisms for platinum introduction and develop processing conditions to eliminate or minimize it.

The first four of these tasks are completed and significant progress has been made on item 5 to make us confident that Pt particles can be eliminated from the laser glass. The primary intent of this paper is to review the currently operating status of Nova and progress on understanding the Pt damage mechanism. The results from other aspects of the work will be briefly summarized to provide necessary background information.

Platinum inclusion damage in laser and optical glasses has been a recurrent problem throughout history of high power lasers. In particular, platinum damage in silicate laser glass has been well known for many years (see, for example, the review in Ref. 3). In some laser systems, the platinum inclusions were not a problem. Our previous high power laser, Shiva, used a Nd:silicate glass and we observed no damage in that system. However, results from current studies of the Nova glass indicate that Shiva operated just below the Pt damage threshold.

It was anticipated that use of a phosphate based laser glass would cause a reduction of the problem with platinum inclusions, because the solubility of Pt is much greater in phosphate glasses than in silicates. When phosphate glass was selected for use in Nova, five 15 cm x 15 cm x 2 cm samples were tested with 1-ns, 1064-nm pulses at 12 J/cm^2 using a 6-cm diameter beam produced by an arm of the Argus laser. These experiments indicated that the glass might typically contain one small (< 10 μm) inclusion in each 10 cm^3. This estimate of inclusion density appears to be correct for all but a few of Nova's disks. However, these qualification studies did not correctly predict the large sizes (up to 250 μm) of some of the inclusions that have been found in glass supplied for Nova, or the growth of the damaged volume that would result from repeated irradiation of those large inclusions.

II. Results from Platinum Inclusion Damage Studies

A. Experimental Studies of Individual Inclusions

Individual inclusions with major dimensions ranging from 4 to 75 μm were tested with 1064-nm pulses having durations of 1.3, 9 or 50 ns. Thresholds for initiation of damage were 2.5 ± .3, 4 ± .4 and 8 ± 1 J/cm^2, respectively, at these three pulse durations. These thresholds scaled with pulse duration (τ) as $\tau^{0.31}$ (Fig. 3) and, as expected for

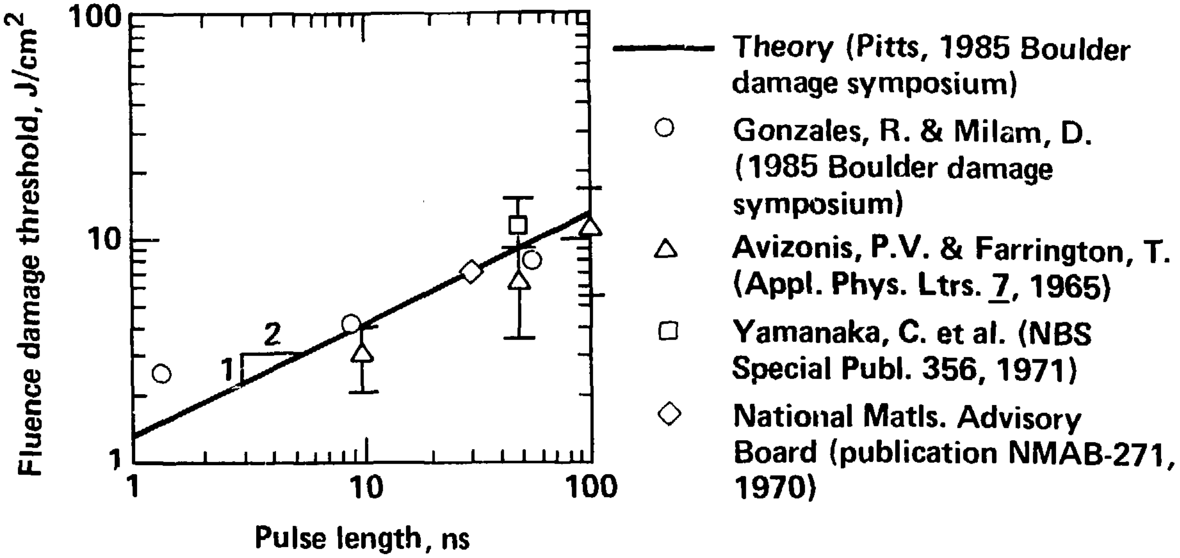

Fig. 3. Measured damage threshold due to Pt° inclusions for laser pulses of several durations. The open circles represent the results from our study and the line represents the threshold calculated by Pitts.[5]

particles greater than one micron, the thresholds were independent of particle size. Damage surrounding small ($\lesssim$ 5 μm) inclusions generally grew to stable dimensions of < 250 μm, while damage surrounding larger inclusions (> 5 μm) grew to dimensions of 400-1000 μm during irradiation by 5-10, 1-ns pulses at 5-9 J/cm^2.[4]

B. Modeling Inclusion Damage

The temperature of a laser-irradiated platinum particle was calculated using a numerical heat transport code (Topaz[5,6]) that allowed heat conduction into the interior of the irradiated particle and into the surrounding glass, and cooling of the particle by radiation. Literature values were used for the absorption coefficient of platinum and for the heats of fusion and vaporization. Temperature dependence of the absorption coefficient was not treated. The Pt vapor state was described using the equation of state employed in the LASEX laser fusion computer code.[7] Details of these calculations are given in the paper by Pitts.[6]

For pulses with duration of 9 or 50 ns, the calculated values of the fluences necessary to heat the surface of the particle to the vaporization temperature of platinum agreed with experimentally measured thresholds for initiation of damage. For 1-ns pulses, the calculated vaporization fluence was about one half of the measured threshold (Fig. 3). Calculated vaporization fluences were independent of particle size (for particles with dimensions > 1.0 μm) and varied with pulse duration as $\tau^{0.5}$. While the discrepancy between calculated and measured 1-ns threshold has not been resolved, there is close agreement between the results of Pitt's calculations[6] and the experimental results obtained both at LLNL and in other studies (Fig. 3). The model calculation suggests that the damage surrounding a platinum particle occurs as a result of fracture driven by the pressure of Pt vaporized from the particle surface. Efforts are underway to model the growth of the damage during repeated irradiation.

C. Status of Nova

A schematic diagram of the components in an arm of Nova is given in Fig. 1. The 12 disk amplifiers are identified by numerals. Figure 2 gives the spatially averaged fluences present in each amplifier during generation of nominal 10-kJ, 1-ns and 20-kJ, 5-ns pulses. Fluence values are those in a plane normal to the beam. Fluences inside a given disk are lower by a factor of n^{-1}, where n is the refractive index of the glass ($n = 1.53$). Also shown are bands representing the measured thresholds for damage to platinum inclusions induced by pulses with durations of either 1 or 5 ns. The displayed thresholds are larger by a factor of 1.53 than the actual measured thresholds (Fig. 3). Recall that the thresholds were measured in samples oriented normal to the test beam. We assume that it would be necessary to increase the beam fluence by a factor of n to damage inclusions inside the Brewster oriented disks.

At the time we began evaluating Nova, each of the ten arms had been used to produce 1-ns pulses with energies in excess of 8 kJ. If the experimental data (Fig. 3) correctly represented the damage thresholds for the glass in Nova, damage should have occurred in each arm in the last three 46-cm amplifiers (numbers 10, 11, 12) and the last 31.5-cm amplifier. Because the local fluence values exceed spatially averaged fluences, some damage might have occurred in the input 46-cm amplifier (#9) and the fourth 31.5-cm amplifier (#7). The 20.8-cm amplifiers and the first three 31.5-cm amplifiers should not have damaged.

Several disks were removed from Nova and inspected. For each of these disks a map was constructed giving the locations and approximate sizes of the damage sites. Similar inspections were made of disks removed from the two-arm laser, Novette, when it was disassembled and moved into place as the final two arms of Nova. Almost all of the 46-cm disks and many 31-cm disks contained damage. The volume density of damages ranged from about 1 to 0.01 cm^{-3}, with typical densities being 0.05 to 0.1 cm^{-3}. None of the 20.8-cm disks were damaged. The sizes of the damage sites depended on the fluences that had been experienced by the disks. This is illustrated in Fig. 4 which gives (for four 46-cm disks) normalized plots of the fraction of sites exceeding a given dimension and in Fig. 5 which gives the median size of damages in seven disks as a function of fluence.

The size of the damages is the parameter that most strongly effects the operation of Nova. Codes that model Nova operation indicate that the beam quality is not seriously degraded if the components contain a low volume density of beam obscurations with sizes < 250 µm. Larger obscurations produce beam ripple that can damage optical elements lying downstream, and very large (>> 1 mm) obscurations block a significant fraction of the beam. Data indicate that disks used at fluences near threshold contained damage smaller than the 250 µm limit. In disks subjected to fluences well above the damage threshold, the median size was greater than 250 µm, and the largest damages had diameters > 10 mm (Fig. 4).

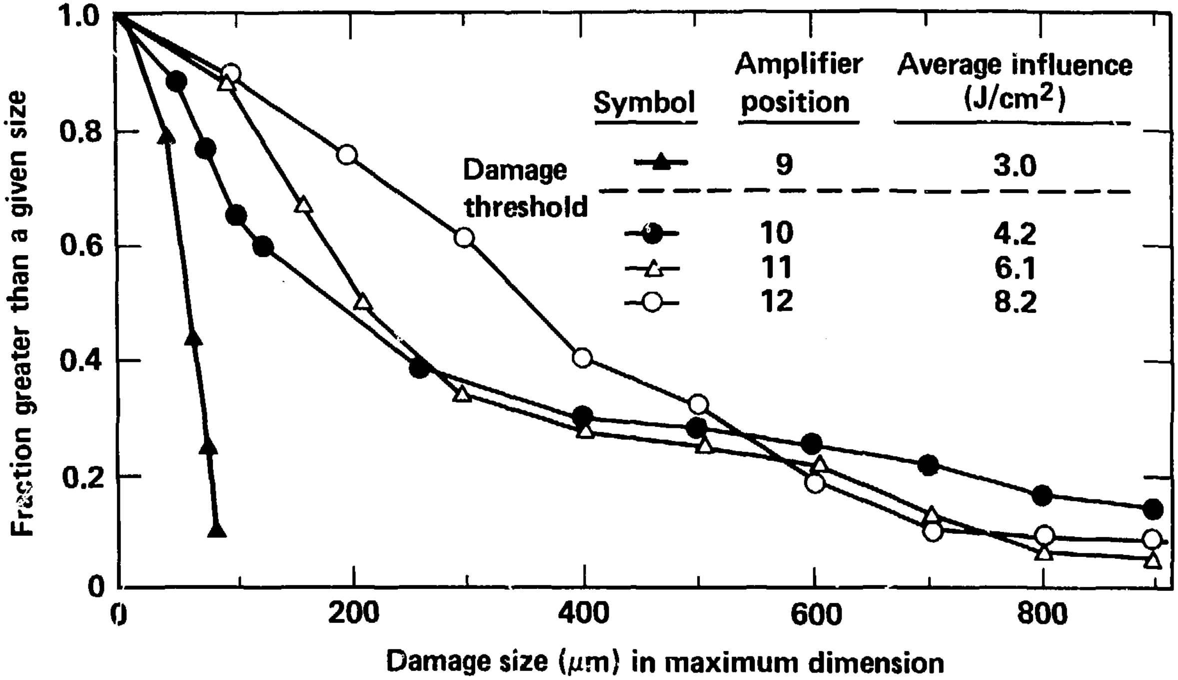

Fig. 4. Normalized plot of fraction of damage sites (in Nova 46cm disks) that exceed the dimension shown on the abscissa. The disk amplifier positions are indicated by the numbers 9, 10, 11 and 12 corresponding to the locations shown in Fig. 1.

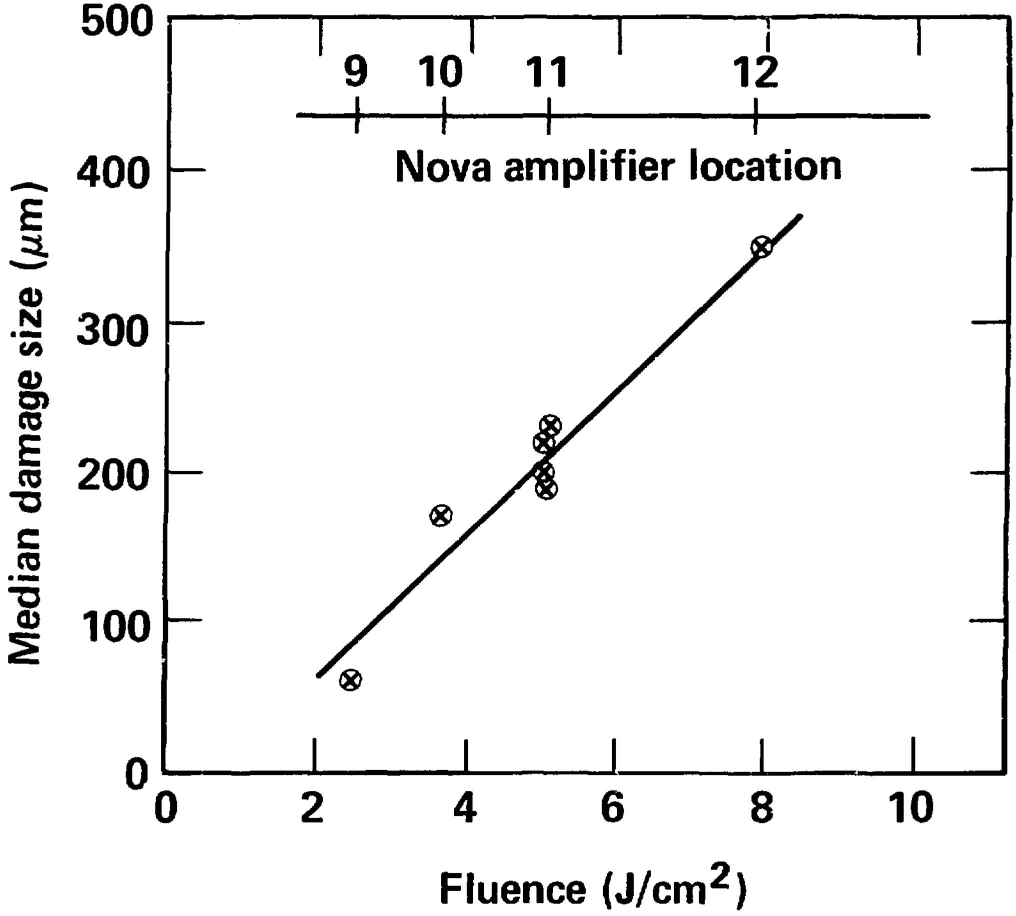

Fig. 5. Median damage size found upon inspection of Nova disks vs. the fluence at that disk location.

The correlation between the level of damage found in several of the Nova disks and the experimental data that we have recently generated can be used to predict which disks are likely to be damaged by a particular use of the laser. For safe operation at 1-ns and 10 kJ per beam (100 kJ total output), replacement of the eight 46-cm disks and of at least eight 31.5 cm disks in each arm is required. If each arm is to safely produce 20 kJ, 5-ns pulses, replacement of almost all 46 and 31.5 disks and the final 20.8-cm disks will be required. Until replacement glass is available, the laser can be safely operated at 5 kJ/arm (50 kJ total) with the existing glass. Currently we have about 2 years of experiments that require less than the 50 kJ output. This is sufficient time for us to replace the current glass with platinum free glass.

D. Inspection of Glass

Certification that Pt° inclusion-free glass has been obtained will require an improved inspection procedure. Previously, the most effective technique for finding a low volume density of inclusions in a large volume of glass was to illuminate the sample from the back or through its edges and visually search for scattering centers. Even after the inclusion problem had become apparent and the visual inspections were being executed with great care, less than 50% of the inclusions were identified in most samples. Detailed microscopic examinations required too much time and were sometimes no more successful. Automated light scattering experiments are probably capable of detecting inclusions, but do not readily distinguish between inclusions and other bulk defects such as bubbles or surface defects. The only procedure known to be completely successful is a destructive damage test. Irradiating an inclusion with 5-10 pulses having fluences well above threshold produces a damaged volume that is readily visible.

To guide the initial experiments aimed at improving glass, two large-area damage experiments were attached to the Nova laser. One was positioned at the midpoint of an arm and allowed irradiation at fluences of 7-10 J/cm^2 of areas with diameters of 6-9 cm. The second was at the output of an arm and allowed similar experiments over areas 25-30 cm in diameter. These experiments have been very successful in determining the damage susceptibility of large glass samples, but use of Nova as a damage tester is highly impractical. To minimize shot time required from Nova, a separate system was built to allow rapid scanning of a glass sample with the beam of a commercially available 20-Hz Q-switched Nd:YAG laser. The beam from the laser is slightly focussed so that the fluence at the sample is about twice the damage threshold for the 8-ns pulses. Successful use of this system to locate platinum inclusions in glass is described in a separate paper by Marion et. al.[8]

E. Progress on Improved Glass Melting

Staff members at LLNL are working with scientists at the two Nova laser glass vendors (Hoya Optic, Inc. and Schott Glass Technologies, Inc.) to eliminate the Pt inclusion problem in phosphate laser glass. Details of glass manufacture at both these facilities are proprietary, so progress can only be discussed in general terms.

In brief, both companies have sought to control the Pt problem by (a) reducing or eliminating possible outside sources of Pt contamination and (b) adjusting process conditions to put or keep all Pt in solution, i.e. as ionic Pt (which does not damage) rather than as metallic inclusions (which do damage).

Considerable progress has been made in both areas. Recently we have tested 7 large pieces of glass having a total volume of about 17-18ℓ and found an average of less than 0.20 inclusions per liter of glass. (Four pieces of glass were completely Pt° free).[8] These new pieces were prepared under conditions designed to convert any metallic Pt° to its ionic form. The samples were tested using both Nova and our new QA inspection tool at 2-4 times the damage fluence. A series of further process changes are in progress that should reduce the Pt inclusion level to less than 0.1 per liter[9] thus meeting our design specifications. For comparison, the current Nova disks have Pt° inclusion concentrations of between 10 to 1000 per liter, thus the current new glass represents a 50 to 5000 fold improvement.

We have begun procurement procedures necessary for the replacement of the Nova glass. Our present plans show construction of production melting facilities beginning in January of 1986 and being completed by about May. Production melting should begin in May or June, and the first replacement glass is expected to arrive at LLNL in Sept. or Oct. The complete replacement process will take about 18 to 24 months.

G. Acknowledgements

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References

1. K. Manes and W. W. Simmons, "Statistical Optics Applied to High-Power Glass Lasers." J. Opt. Soc. Am. A 2, 1985, pg. 528.
2. J. T. Hunt and R. J. Foley "Laser Systems and Operations" in Laser Program Annual Rept, UCRL-50021-84, LLNL, (1985).
3. "Fundamentals of Damage in Laser Glass" National Materials Advisory Board, Division of Engineering, National Research Council; report NMAB-271, Washington, D. C. July 1970.
4. Ray Gonzales and Dave Milam "Evolution During Multiple-shot Irradiation of Damage Surrounding Isolated Pt Inclusions in Phosphate Laser Glass." 17th Boulder Damage Symposium, Oct. 1985.
5. A. B. Shapiro "Topaz - A finite element heat conduction code for analyzing 2-D solids" UCID-20045, LLNL, March, 1984.
6. J. H. Pitts "Modeling laser damage caused by platinum inclusions in laser glass", 17th Boulder Damage Symposium, Oct. 1985, Boulder, CO.
7. C. Orth, Lawrence Livermore National Laboratory, Livermore, CA, Private Communication, July 1985.
8. J. E. Marion, P. H. Chaffee, G. J. Greiner and J. H. Campbell "A versatile laser glass inspection and damage testing facility", 17th Boulder Damage Symposium, Oct. 1985, Boulder, CO.
9. C. W. Hatcher, LLNL, 1985, unpublished results.

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