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DAMAGE RESISTANCE OF COATED OPTICS

by

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INTRODUCTION

Successful application of high energy lasers to national needs in strategic defense, inertial confinement fusion and isotope separation is contingent on the development of optical coatings that can withstand very intense radiation. Significant progress has been made in the development of damage resistant coatings over the past 15 years to support the needs of the inertial confinement fusion and isotope separation programs. However, the requirements evolving from the Strategic Defense Initiative (SDI) are much more severe and demand dramatic improvements in the performance of optical coatings beyond the current state of the art. Today I will present the approach that we are taking at Los Alamos towards solving the problems of optical coatings for SDI.

An Optical Damage Program has been set up at Los Alamos to develop damage-resistant optical coatings for high-power visible and ultraviolet lasers in strategic defense, inertial confinement fusion and special isotope separation. For strategic defense the program is addressing ground-based lasers (GBL), specifically, the rf LINAC free-electron laser (FEL) being developed by Los Alamos and the XeF laser, Raman-shifted in H_2 (EMRLD), being developed by AVCO and Rocketdyne. For inertial confinement fusion we are supporting the development of the KrF laser at Los Alamos as a fusion driver. Finally, in the special isotope separation program we are developing optical coatings for Alexandrite lasers. Since the eventual goal of the FEL program is a laser around 0.5 to 1.0 μm, the Optical Damage Program at Los Alamos is concerned with the entire wavelength range between 0.25 and 1.0 μm.

ISSUES FOR GBL OPTICS: INTEGRATED EFFECTS

The issues associated with GBL optical coatings is being addressed with a joint program funded by the Directed Energy Weapons Office of the SDIO and involving the Naval Weapons Center (NWC), the Air Force Weapons Laboratory (AFWL), and Los Alamos. Overall program management is under the direction of James Stanford of China Lake (NWC). China Lake also has responsibility for a portion of the fundamental research on laser-material interactions. The Air Force Weapons Laboratory has responsibility for the development of novel coating deposition techniques and for optical and mechanical characterization of coatings. Los Alamos has responsibility for optimization of conventional coating techniques, such as thermal and electron-beam evaporation, for damage testing, for integrated effects and for further research on laser-material interations.

The optics located in the laser cavity represent the area of highest risk for GBL's, because the optics are exposed to damaging environmental effects other than the fundamental laser radiation. Figure 1 lists some of the multiple hazards for both the excimer laser and the FEL. Considerable data already exists on the degradation of windows and/or mirrors on an XeF laser that arises from exposure to the corrosive laser gas mix which contains fluorine. The reactivity of the gas mix is enhanced when it is energized by a high voltage electron beam. The latter also generates X-rays and scattered electrons which irradiate the optical components. Finally, although it is not evident that exposure to high pressure H_2 in the Raman cell will cause problems, the data base is inadequate to assess the performance of optical coatings in this environment.

Comparable hazards exist for FEL's which extract light from very high energy (100 MeV), relativistic electron beams. The electron beam generates γ-rays through Bremsstrahlung. Additional γ-rays as well as neutrons are produced when the electron beam is collimated by passing through a scraper or when the electrons are stopped in a beam dump at the end of the accelerator. The FEL optics will be exposed to significant amounts of ionizing radiation, both γ-rays and neutrons. The optics inside the FEL will reside in vacuum, which can effect the performance of the coatings. Finally, the FEL generates radiation at the higher harmonics of the fundamental laser frequency. For FEL's operating in the 0.5-1.0 μm wavelength range, it does not take many harmonics before the coatings will become strongly absorbing.

The amount of radiation generated at higher harmonic frequencies depends strongly on the characteristics of the FEL. For example, the second and third harmonics were down in intensity by 10^5 for the Los Alamos FEL which operated at 10 μm, while the third harmonic was down by less than 3 for Orsay FEL which operated at 0.6 μm. For visible or near visible FEL's the energy in the higher harmonics, even if very low, will be absorbed and may create color centers which in turn absorb at the fundamental laser frequency, thereby creating a major problem for the optical coatings. This is an important area for FEL's which must be addressed.

The multiple hazards for the laser cavity optics must be considered for possible synergisms not just as separate, independent sources of damage or degradation. One possible synergism, the creation of color centers by higher harmonics or ionizing radiation, has already been mentioned. Another possibility would be electron or photon (X-ray) stimulated desorption altering the surface of a coating in an XeF laser so that the fluorine resistance was degraded. Experiments at Washington State University (WSU) and at the University of Rochester Laboratory for Laser Energetics (LLE) have already demonstrated enhanced erosion of optical coatings exposed to both fluorine and electrons.

The environmental hazards severely limit the choice of materials that can be considered for optical coatings. First, to insure minimal absorption of the intense visible and ultraviolet laser radiation one must use large band gap dielectrics such as metallic fluorides and refractory oxides. For wavelengths longer than 0.5 μm one can also consider metal mirrors at grazing incidence. Even here the choice is limited to silver for visible lasers.

Second, one must consider the effects of ionizing radiation such as X-rays, γ-rays, etc. In this context metals are far superior to dielectrics since they do not suffer ionization damage. However, both dielectrics and metals are susceptible to displacement damage that can arise from exposure to energetic neutrons. Among the dielectrics, radiation damage experiments have shown that oxides are generally far superior to fluorides, especially alkali fluorides.

Finally, one must consider the chemical environment. For excimer lasers the coatings will be exposed to molecular fluorine (F_2), fluorine atoms (F), and fluoride ions (F^-). Our experience to date indicates that alumina (Al_2O_3) and cryolite (Na_3AlF_6) give the best fluorine resistance for short wavelength coatings. Materials such as magnesium fluoride (MgF_2) and silica (SiO_2) exhibit very poor fluorine resistance. For FEL's the coatings will be in vacuum so that one should avoid certain oxides, such as titania (TiO_2) which tend to lose oxygen under vacuum.

ISSUES FOR GBL OPTICS: DAMAGE RESISTANCE REQUIREMENTS

To illustrate the types of improvements in coatings required by GBL's for strategic defense, let us consider a specific example from the EMRLD program. Similar problems will arise for FEL's as the designs mature and solidify. At present the optical component, which appears to be under greatest stress in the EMRLD design, is a fluorine-resistant, anti-relective coated calcium fluoride (CaF_2) window on the laser power amplifier. The current design calls for a damage threshold of 9 J/cm^2. Rocketdyne has procured several small coated parts from two commercial vendors and sent the parts to Los Alamos for damage testing. The first part we tested exhibited catastrophic damage at fluences comparable to the design requirement. However, microscopic damage was <u>always</u> observed, down to 1.7 J/cm^2. In catastrophic damage the damage spot continues to grow until it eventually fills the entire footprint of the laser beam. In microscopic damage

5-20 μm diameter pits are formed, but they do not grow with subsequent laser shots. The problem is that the relationship between microscopic damage and macroscopic changes in reflection or transmission is not known. Extensive microscopic damage could significantly degrade the performance the laser cavity windows. More serious could be the effect on fluorine resistance, which could be reduced by the microscopic damage pits. Experiments are planned to elucidate the effect of microscopic damage on the optical performance and chemical stability of the windows.

Damage thresholds for other small parts provided through Rocketdyne have ranged between 2-4 J/cm^2 with the catastrophic and microscopic thresholds being comparable. Therefore, the commercially coated parts have damage thresholds two to three times lower than the design value. Moreover, we have not included any design margin nor have we accounted for any degradation in performance of the coated optical component on scaling from two inch diameter test samples to the large windows required for a high-power GBL.

More extensive testing of fluorine-resistant, anti-reflective coatings has been performed for the KrF fusion laser development program by Stephen Foltyn's group at Los Alamos. Based on these results and assuming a factor of two improvement, which has generally been observed from 248 nm to 351 nm, the current state of the art is about 5 J/cm^2 for coatings needed in strategic defense applications. This estimate, however, does not account for one important fact, namely, that the laser pulse length for EMRLD will be around 1 μs, while our damage testing is performed with pulse lengths of 10-20 ns. The information on scaling damage thresholds with pulse length is extremely scarce, but the initial data taken by Foltyn (Los Alamos) and Marrs (NWC) indicates that the damage threshold may be a factor of two higher for the EMRLD pulse length. Incorporating the pulse length scaling into our latest test results yields damage thresholds approaching the design requirements. It appears we are close, but we need much more detailed information on scaling with pulse length and with the size of component and on the effect of microscopic damage.

APPROACH TO MEET GBL REQUIREMENTS

The approach we are taking to develop coatings that meet the requirements for GBL's may be divided into three phases. The first phase consists of identifying the best materials for optical coatings as well as identifying the commercial vendors that achieve the best results with these materials. An iterative Edisonian survey, in which samples are procured from industry, tested at Los Alamos and a new set procured, can yield the most promising candidates very efficiently.

The second phase, into which we are currently entering, is concerned with optimization and performance validation of the materials and vendors identified in phase one. Quality assurance will be essential for designing and building large GBL's. A cooperative R&D program will be initiated between Los Alamos and a few selected commercial coating companies. The purpose will be to combine industrial expertise in coating fabrication with expertise in damage phenomenology and materials science at Los Alamos in order to optimize coating performance.

It is apparent that we must make dramatic improvements or breakthroughs in the quality of optical coatings to satisfy the requirements of strategic defense. We are still far from the intrinsic damage limits of optical coatings. Much more fundamental research on the mechanisms of laser-induced damage is required to realize the potential performance of coatings. The key is to relate damage mechanisms and thresholds to microscopic thin film properties and, in turn, to relate thin film properties to deposition parameters. Achievement of this goal will require strong participation from industry, universities and a wide range of federal laboratories.

CONVENTIONAL COATING OPTIMIZATION

Figure 2 illustrates the improvement in damage resistance that can be made by identifying the appropriate materials and vendors. The data were taken from the initial development phase of optical components for the KrF fusion laser program. The progress achieved in three years is impressive, passing from microscopic damage thresholds to thresholds greater than 6 J/cm^2, which exceeds design requirements.

Multilayer dielectric coatings of alumina (Al_2O_3) alternating with silica (SiO_2), which yield the best results for reflectors at 248 nm, are also prime candidates for reflectors at 351 nm. However, as shown dramatically in Figure 3, the scatter in performance of alumina/silica coatings from different vendors and even from the same vendor is tremendous. For example, vendor A delivered a coating with a damage threshold approaching 15 J/cm^2, which exceeds the requirements of EMRLD, but they also delivered samples with thresholds of 6-7 J/cm^2, which do not meet the requirements. Even more startling is the range of thresholds from 1 to 8 J/cm^2 obtained for coatings from vendor E. Both the need for performance validation and the promise for performance optimization are evident. One final point, most of the coatings were produced using conventional electron-beam deposition. The one sample produced using a novel deposition technique, ion-beam sputtering, gave a low damage threshold (1 J/cm^2). This illustrates the importance of a concerted effort to optimize conventionally deposited coatings.

The scatter in performance observed between sets of small (two-inch diameter) samples is also observed across the surface of large optical components. Figure 4 shows measurements made by Stephen Foltyn at Los Alamos on an 8"-square coated with alumina/silica to reflect 248 nm. The square was scribed into 16 two-inch square segments, which were then tested for damage independently. The average of the sixteen measured damage thresholds (4.2 J/cm^2) was close to the values obtained from three two-inch samples coated earlier by the same vendor. However, the damage thresholds on the 8"-square varied from 2.0 to 5.5 J/cm^2. Clearly the coating is not uniform and there are different types of defects in the coating, some of which have low damage thresholds. This illustrates a major problem in quality assurance for large coated optics. Thousands of spots on the coating had to be tested to locate a few damage prone areas. However, the latter determine the damage resistance of the component, since the weak areas will be found as soon as the coating is irradiated with a large spot laser. In this case, the actual damage threshold is a factor of two lower than the values measured on two-inch samples, which illustrates the problems of scaling to large optics.

So far we have only considered coated optics located in a "benign" environment, i.e., exposure to intense laser radiation is only the concern. As discussed earlier, laser cavity optics are exposed to multiple hazards. For example, XeF and KrF cavity optics must be fluorine resistant. Alumina/silica multilayer dielectric coatings have demonstrated the best damage resistance, but silica is not fluorine resistant and must be replaced for cavity optics. We have identified one promising fluorine resistant combination, namely, alumina/cryolite (Na_3AlF_6). A simple test for fluorine resistance is static exposure to a mixture of 0.6% F_2 in He, which simulates the laser gas mix. Figure 5 shows the effect of static fluorine exposure on reflectance at 248 nm for three alumina/cyrolite mirrors obtained from different vendors. Only one coating retains high reflectance after 100-200 hours of exposure.

To optimize performance in XeF the laser gas is actually heated to about 150°C. We have built a cell which enables us to test for damage in the presence of hot fluorine. Our preliminary results indicate that there is not much difference between the damage thresholds measured in air and in hot fluorine for coatings that do well under the static fluorine exposure. This result greatly facilitates testing of fluorine resistance.

NOVEL COATING TECHNIQUES

Another approach to achieve the damage resistance required in strategic defense applications is to investigate alternate or novel techniques for depositing optical coatings. One method which has already shown great promise in yielding high damage thresholds is chemical deposition using sol-gels. The development of sol-gel coatings has been advanced most recently by the Lawrence

Livermore National Laboratory (LLNL) in conjunction with laser fusion. The first samples we received from LLNL for testing displayed high damage resistance. Damage thresholds of 5 J/cm^2 were measured at 248 nm for two anti-reflective silica sol-gel coatings on fused silica. During the tests of the front surface the back surface was being irradiated at subthreshold fluences. Many workers, including ourselves, have observed that subthreshold irradiation can "harden" an optical coating, actually enhancing the damage resistance. As a test of this phenomenon, the back surface was tested and a threshold of 14 J/cm^2 was measured! As a further test, the sample was allowed to sit for three months and then was tested again, giving a threshold of 9 J/cm^2. One drawback of silica sol-gel coatings, however, is the lack of fluorine resistance. Nevertheless, sol-gels and laser hardening are two items which need to be pursued vigorously in the future.

LOS ALAMOS OPTICAL DAMAGE PROGRAM

Los Alamos has put together a broadly based optical damage program to develop damage resistant optical coatings for high-power visible and ultraviolet lasers. The basic elements of the program are conventional coating fabrication, damage testing, novel coating fabrication and materials analysis. A brief description of each aspect of the program will be presented below.

Industrial participation is the key to conventional coating fabrication. As indicated above, contracts will be let under the auspices of the SDI GBL coatings program to two or more commercial coating manufacturers. The purpose will be to optimize the performance of current coating technology through a joint R&D effort which combines industrial coating expertise with damage testing and materials science at Los Alamos. Initially the emphasis will be placed on developing fluorine-resistant, damage-resistant, anti-reflective coatings for high-power XeF lasers.

Los Alamos possesses extensive capabilities for damage testing of optical components. There are two damage test laboratories dedicated to XeF, one dedicated to simulate FEL's at 1.0 and 0.5 μm, one dedicated to XeCl and XeCl pumped dyes, and one shared between KrF and Alexandrite. Scientists at Los Alamos have pioneered the use video equipment to monitor damage testing, which allows efficient testing of hundreds of sites each tested with hundreds of laser shots. This large amount of data when coupled with new techniques for analysis yields accurate damage thresholds.

Integrated effects provide the major uncertainty in the performance of coated optics for GBL's because the data base is extremely sparce. Therefore, a major emphasis at Los Alamos will be to characterize the multiple hazards in laser cavities that give rise to integrated effects. Schmid (LLE) and Dickinson (WSU) have already shown that there is a significant synergism between fluorine and electrons in etching optical coatings. We plan to augment the ongoing fluorine testing, which was described earlier, with a detailed study of the surface chemistry of F_2 and F with coatings using molecular and atomic beams and sophisticated surface analysis.

Optics in both of the proposed GBL's, XeF and the FEL, will be exposed to ionizing radiation. We plan to use the accelerator at the Los Alamos Meson Physics Facility (LAMPF) as well as a Co^{60} source to irradiate coated optics and simulate the neutron and γ-ray flux expected in the FEL. Measurements will also be made in one of the high power KrF lasers, built for the fusion program at Los Alamos, to determine the X-ray and electron flux expected on optics in the EMRLD XeF laser. At present there are no data on the dose of ionizing radiation at cavity optics for high power, electron beam pumped excimer lasers.

The mechanisms of laser-induced damage are not understood, although they are known to depend on materials, wavelength, pulse length, etc.. Present day coatings are riddled with defects, but not all defects lower damage resistance. A key to illuminating the phenomenon of laser-induced damage will be the development of nondestructive probes that can identify damage prone areas in optical coatings and thereby enable detailed materials analysis to determine the type of defect that is reducing the damage resistance. Two promising defect identification techniques have been developed recently: (1) laser scattering by Marrs at NWC and (2) photothermal spectroscopy by Schmid at LLE. A major element in the damage phenomenology effort at Los Alamos will be to devise additional defect identification techniques as well as to refine the existing ones. We also plan to initiate a significant theoretical effort on laser-induced damage in cooperation with John McIver at the University of New Mexico, building on our strength in chemical dynamics and his strength in laser-material interactions.

As discussed earlier the optics in excimer laser cavities may be exposed to significant doses of electrons and X-rays. The energetic electrons and X-rays may damage or degrade the optical coatings by stimulating desorption through electronic excitation. We are initiating a project with Norman Tolk at Vanderbilt University to investigate this possible damage mechanism. Tolk has previously shown that neutral atoms are orders-of-magnitude more efficiently desorbed than ions. Rothenberg and Kelly at IBM have recently observed significant desorption of Al_2O_3 upon irradiation at 0.25 μm with 0.5 J/cm^2. Electron and photon stimulated desorption may be an important damage mechanism in high power excimer lasers.

Another major thrust of the program is applying sophisticated materials analysis to correlate thin-film properties with damage resistance. In particular, we will be employing ion-beam analysis to determine stoichiometry, density and impurities as a function of depth in multilayer optical coatings. We will also be employing more conventional surface analytical techniques such as Auger electron spectroscopy, electron microscopy, ion scattering, etc.. Finally, in conjunction with Angus Macleod at the University of Arizona we will be developing theoretical models of thin film growth.

The last element of the Los Alamos Optical Damage Program is novel coating fabrication. We have contracts with Angus Macleod at the University of Arizona and Robert McNeil at the University of New Mexico to study ion-assisted deposition of optical coatings. We are also investigating novel metal coatings at Los Alamos in support of our FEL program.

SUMMARY

In summary, the objective of the Optical Damage Program at Los Alamos is to develop damage resistant coatings for high power visible and ultraviolet lasers in applications to strategic defense, inertial confinement fusion and special isotope separation. Significant progress is being made as attested by the successful operation at Los Alamos of the KrF Large Aperture Module, which produced 10.5 kJ in a single pulse. The optical coatings performed as required. This is encouraging in light of the much more severe requirements on optical coatings posed by GBL's for strategic defense. The key to our achieving these new ambitious goals will be a strong, concerted effort on understanding the basic mechanisms of laser-induced damage. This is an exciting challenge to which we look forward.

MULTIPLE HAZARDS FOR LASER CAVITY OPTICS

EXCIMER

- FUNDAMENTAL RADIATION
- X-RAYS
- ELECTRONS
- CORROSIVE GASES (F_2, F, F^-)
- HIGH PRESSURE H_2 (RAMAN CELL)

FEL

- FUNDAMENTAL RADIATION
- γ-RAYS
- NEUTRONS
- VACUUM
- HARMONICS

POSSIBLE SYNERGISMS:

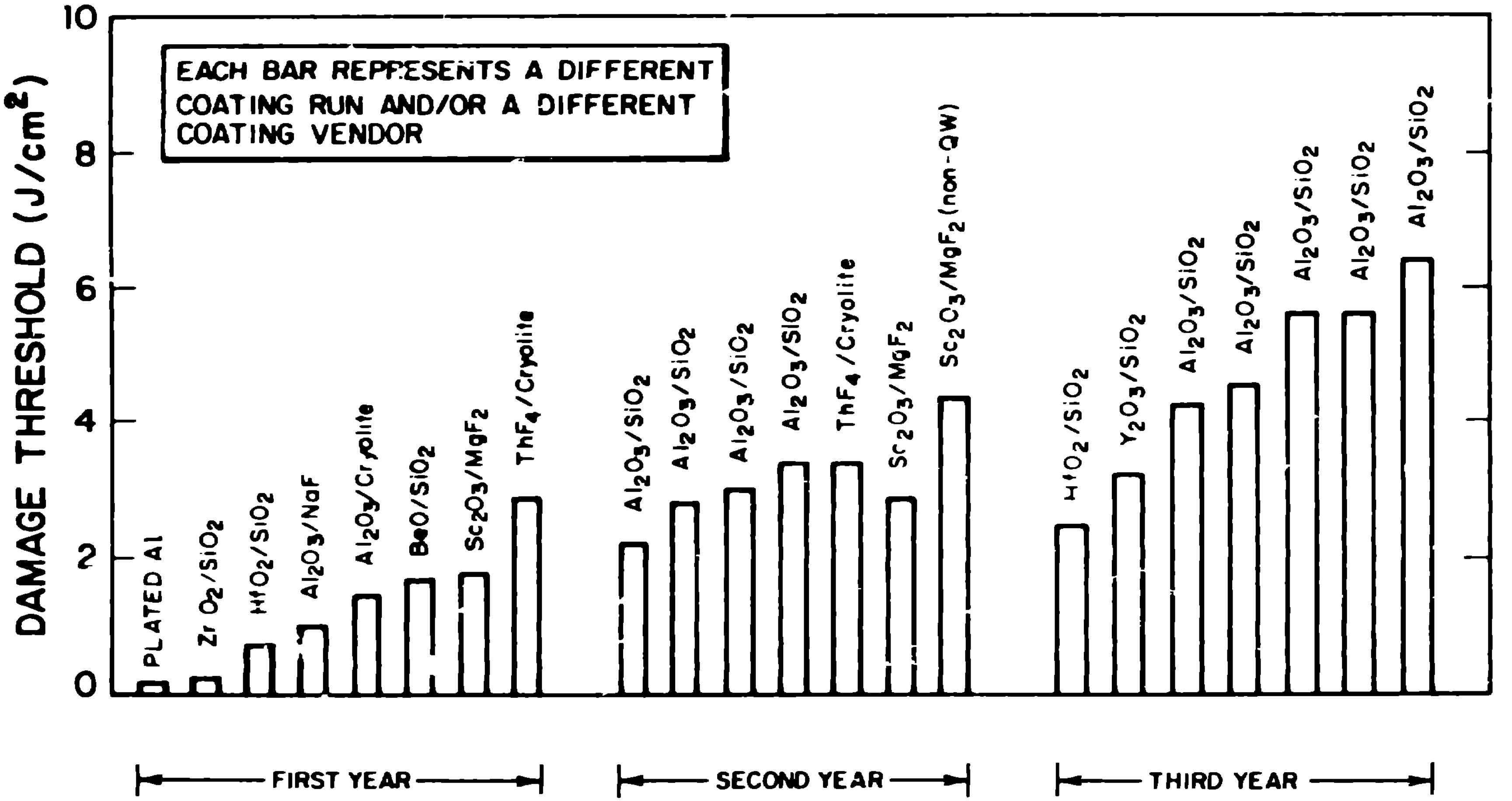
- IONIZING RADIATION → COLOR CENTERS → ABSORPTION OF FUNDAMENTAL
- ELECTRON AND PHOTON STIMULATED DESORPTION → HIGHER FLUORINE REACTIVITY

CHM -VG -7507

Figure 1

OPTIMIZATION OF 248 nm REFLECTORS

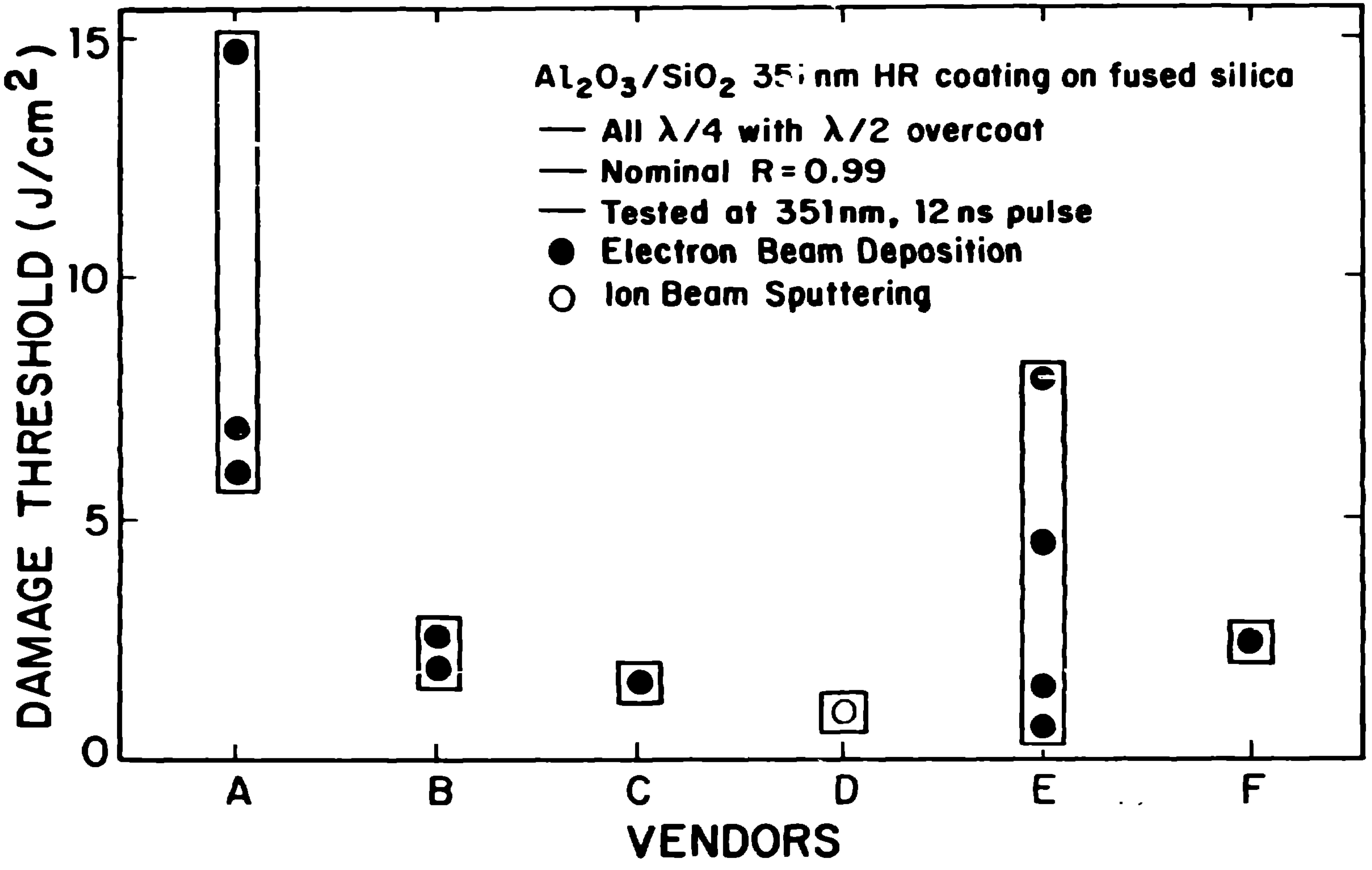
Figure 2



AP-1-VG-8500A

LARGE SCATTER IN DAMAGE RESISTANCE OF COMMERCIALLY COATED OPTICS

Figure 3



CHM-VG-7522

SPREAD IN PERFORMANCE EVIDENT IN SCALING TO LARGER OPTICS

- DAMAGE TEST OF $A\ell_2O_3/SiO_2$ COATING OF 8" SQUARE REFLECTOR
  - SAME VENDOR COATING TESTED EARLIER ON 2" DIAMETER SAMPLES
  - DAMAGE THRESHOLDS MEASURED: 3.6, 3.8, 4.2 J/cm^2
- SCRIBED 8" OPTIC INTO 16 2" SQUARES
  - INDEPENDENT BUT IDENTICAL TESTS OF 2" SQUARES
- RESULTS (MEASURED THRESHOLDS INDICATED IN J/cm^2)

3.0	4.0	2.0	5.5
3.5	5.5	2.0	4.0
5.5	5.5	5.5	4.5
4.0	5.5	3.5	4.0

AVERAGE THRESHOLD
4.2 J/cm^2

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Figure 4

KrF CAVITY OPTICS ARE EXPOSED TO CORROSIVE F_2

- BEST DAMAGE THRESHOLDS WITH Al_2O_3/SiO_2
  - Al_2O_3 FLUORINE RESISTANT, SiO_2 IS NOT
  - REPLACE SiO_2 WITH CRYOLITE (Na_3AlF_6)
- TEST F_2 RESISTANCE BY STATIC EXPOSURE TO 0.5% F_2 IN He
  - TO DATE DAMAGE TESTS IN F_2 YIELD COMPARABLE THRESHOLDS TO TESTS IN AIR

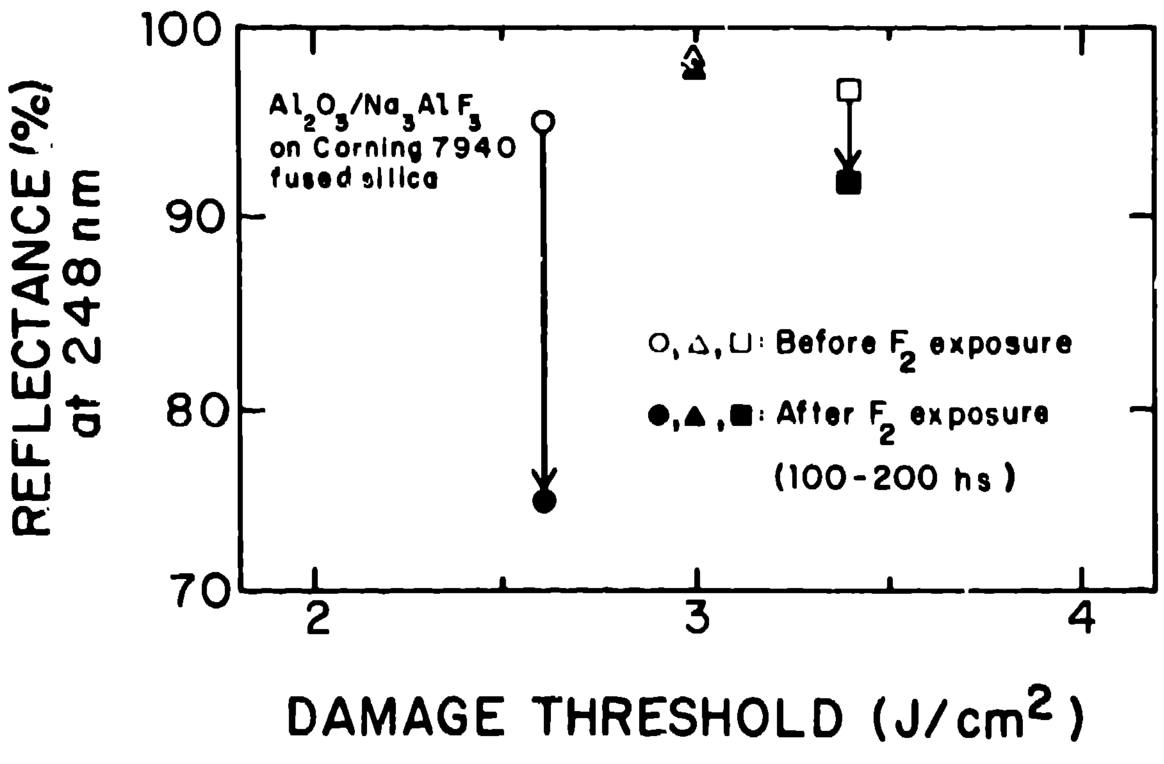

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Figure 5