2

Conf-9010287--3

UCRL-JC-104970
PREPRINT

JAN 1 6 1991

High Threshold HR Coatings at 1064 nm

F. Rainer, F. P. De Marco,
J. T. Hunt and A. J. Morgan

This paper was prepared for submittal to
The 1990 Boulder Damage Symposium
Boulder, Colorado
Oct. 24-26, 1990

December 17, 1990

Lawrence Livermore National Laboratory

DISCLAIMER

UCRL-JC--104970

DE91 006236

High threshold HR coatings at 1064 nm

F. Rainer, F. P. De Marco, J. T. Hunt and A. J. Morgan

Lawrence Livermore National Laboratory
P.O. Box 5508, L-490, Livermore, CA 94550

and

L. P. Mott, F. Marcelja and M. R. Greenberg

Spectra-Physics Optics Corporation
1330 West Middlefield Road, Mountain View, CA 94039-7013

ABSTRACT

We have conducted an extensive series of laser damage measurements on highly reflective (HR) dielectric coatings which have yielded 1064-nm thresholds as high as 40 J/cm^2 for 8- to 10-ns pulses at pulse-repetition frequencies (PRF) of 10 Hz. Moreover, by laser conditioning these coatings with sub-threshold pulses, the thresholds of some coatings were raised to levels exceeding 70 J/cm^2. These are the highest threshold dielectric HR coatings that we have tested in this regime.

The coatings were originally developed to produce HR-overcoated metal mirrors for free-electron-laser (FEL) applications at high PRF. Our tests included coatings deposited on both dielectric substrates and molybdenum (Mo) substrates. In each category we also examined coatings with a pre-coat of Mo between the substrate and the HR stack. The improved dielectric HR stacks effectively shielded the Mo from the laser irradiation so that the thresholds of virtually all Mo samples exceeded levels of the best dielectric-enhanced and dielectric-HR-coated metal mirrors we have tested to date.

In addition to the low PRF measurements, we also conducted 1064-nm damage tests at 6-kHz PRF using 65-ns pulses from the Kilroy damage test facility. The coatings survived thermal loading of fluences ranging from 2 to 10 J/cm^2 with respective small spot sizes on the order of 1.2 to 0.3 mm (1/e^2 diameter).

1. INTRODUCTION

We have endeavored to improve laser damage thresholds of HR coatings at LLNL both in support of the inertial confinement fusion (ICF) and FEL programs. These efforts have involved several commercial vendors applying techniques to improve coating designs, deposition techniques, substrate preparation methods, cleaning techniques, choice of coating materials, and post-deposition processing. The current work reflects the results of a collaborative effort between Spectra-Physics Optics Corporation (SPOC) and the Lawrence Livermore National Laboratory (LLNL) to apply some of these techniques in order to develop high threshold, e-beam-deposited, dielectric HR coatings suitable for deposition on metal substrates.

The primary motivation for depositing HR coatings on metal substrates was to provide an efficient avenue for cooling the coatings when subjected to low level absorption at high PRF's. This is important for FEL applications. However, we have found that some of the coating, substrate preparation, and post-deposition techniques yielded HR coatings on dielectric substrates with some of the highest laser damage thresholds we have measured to date. These results are particularly beneficial for use in ICF lasers under single-shot or low PRF operating conditions.

Although not all permutations of design and operating parameters were attempted, we determined that several promising techniques were worthwhile considering in producing high threshold coatings. These included:

i. Quarter-wave and non-quarter-wave layer designs using hafnia and silica as the constituent materials.
ii. Composite substrates consisting of multiple thick layers of stress-balanced alumina and silica which were deposited on the substrate and polished before the final HR deposition.
iii. Laser conditioning of the coatings by irradiating them in a ramped fashion with multiple shots of gradually increasing fluence.

2. LOW PRF COATING DEVELOPMENT CHRONOLOGY

2.1. Initial experiments with HR-overcoated Mo substrates

In 1988 our first attempts at fabricating e-beam-deposited, dielectric HR coatings on Mo substrates yielded only marginal results. Cosmetically the samples looked quite good, but under 100x or greater magnification, Nomarski microscope photographs showed an abundance of polishing scratches and defects ranging from < 1- to 20-µm diameter. The scratches were essentially on the metal rather than the HR stack. The other defects were most probably also at the substrate-coating interface. The density of defects ranged from a few to > 100/mm^2, but only a very small fraction of them actually contributed to the onset of damage at threshold fluences. We attributed the scattered test results to the variation in defect density. We found sites (~ 1 mm^2) that survived fluences as high as 19 J/cm^2 when irradiated with 16-ns pulses at 1064 nm. However, by scanning larger areas (> 1 mm^2) on the best sample we measured a threshold of only 3.4 J/cm^2. All threshold measurements reported in this work were measured to an uncertainty of ± 15%.

In general we found that these samples looked and behaved about the same as other metal mirrors that we had tested earlier and they fell within the upper range of damage thresholds reported in our database.[1] Our highest threshold to date had been at 4.3 J/cm^2. Typically, we observed an improvement of about a factor of two in thresholds over comparable bare metal mirrors without either a dielectric HR or at least a single-layer dielectric overcoat. We attributed the low-fluence failure of these overcoated metal mirrors to two factors. (1) When a dielectric HR stack was deposited on the metal surface, macroscopic-sized (5 to 100 µm) defects on the substrate printed through the first and successive dielectric layers so that the coating thickness at these defect sites differed slightly from that of the surrounding flat area. This anomaly had the effect that the stack at the defect locations was not a true HR. (2) Alternatively the HR stack may have had pinholes through some or many layers. The net effect would be that in either case some high fluence light could propagate through the entire dielectric stack. For transparent dielectric substrates such light energy would leak through the substrate. However, for metallic substrates, whatever energy was not converted directly to heat by absorption at the interface would be reflected back and be trapped by the HR stack. Effectively this trapped energy could force the stack to rupture. The problem would continue to be exacerbated with successive pulses in a PRF mode of irradiation.

2.2. Task I experiments of HR coatings on dielectric and Mo substrates

We subsequently had fabricated a set of Mo substrates to be polished on a best-effort basis by United Technologies Optical Systems (UTOS). A representative sample was measured to have an RMS roughness ranging from 15 to 18 Å at three sites. SPOC then fabricated the following matrix of dielectric HR coatings:

i. Three types of substrates — polished Mo, fused silica, and silicon.
ii. With and without a precoat of Mo directly on the bare substrate before the HR coating was deposited.

iii. Three dielectric HR coating designs with the following material combinations — zirconia/silica, titania/silica/hafnia, and tantala/silica.

The Mo precoat was evaporated onto the substrates with an e-beam-source technique developed by SPOC. This was done to assess the effect of the difference between the rougher Mo substrate surface and the smoother evaporated Mo layer. As an alternative to a Mo substrate we fabricated some coatings on silicon substrates because silicon has a high thermal conductivity and can be polished to a much smoother surface figure than the bare Mo.

LLNL conducted damage tests on these samples using the Reptile damage test facility. The samples were irradiated at 1064 nm with 10-ns pulses at 10-Hz PRF. Each site was irradiated with a nominal Gaussian beam profile ≥ 1.0-mm diameter ($1/e^2$) with at least 600 shots at the same fluence unless massive damage ensued earlier. The test results are shown in Fig. 1 which was presented as an example of the status of overcoated metal-mirror damage thresholds at the 1989 Boulder Damage Symposium.[2]

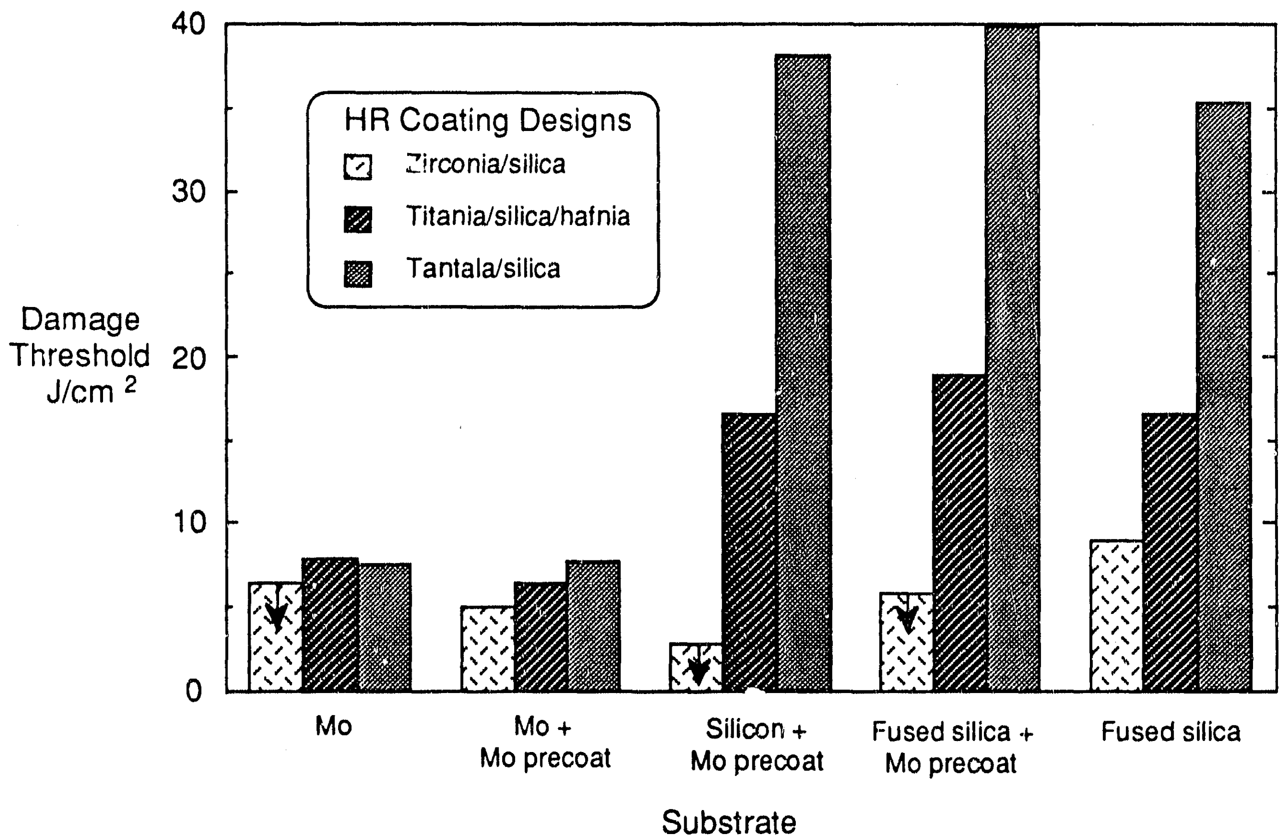

Fig.1. Unconditioned laser damage thresholds of three coating designs, on three substrate materials, with and without evaporated Mo precoats between the substrates and HR coatings (SPOC — Task I). The thresholds were measured with 600 pulses per site using Gaussian spot diameters ≥ 1 mm ($1/e^2$) at 1064 nm with 10-ns pulses at 10-Hz PRF. The arrows indicate that the thresholds for those particular samples were less than the minimum test fluence shown. All samples were prepared in a small production chamber.

We drew several general conclusions from this first round of tests:

i. All of the samples with Mo substrates had low thresholds ranging from 5 to 8 J/cm^2 because of a high density of substrate defects which printed through both the optional Mo precoat and the HR stack. However, we found that most of these thresholds were higher (as much as almost a factor of two) than those previously measured as noted above. Although some of this improvement was attributed to a better polish on the substrates, we feel that the improved HR coatings shielded the substrates better from light leakage to the substrate-coating interface.

ii. With a given HR stack design there was essentially no difference in thresholds for any non-Mo substrate. This included those samples with an evaporated Mo precoat between the substrate and the HR coating. There was more scatter in the zirconia/silica data because these coatings had a higher density of pre-irradiation defects.

iii. The thresholds ranked as follows for the coatings not on Mo substrates:

tantala/silica	35 - 40 J/cm^2	clean looking
titania/silica/hafnia	16 - 19 J/cm^2	clean looking
zirconia/silica	< 9 J/cm^2	many visible defects.

The damage morphology in most cases was typical of what we had observed previously in HR coatings. A small fraction of pre-irradiation defects, usually 5 to 10 µm in size, provided initiation sites for damage. A 100- to 500-µm circular area centered about the damage-causing defect delaminated, often without any perceptible change in the defect itself. These delaminations were virtually impossible to detect with Nomarski microscopy because the delaminations remained intact. They were observed with bright-light illumination at 100x magnification. Occasionally a defect may have been slightly altered in appearance or size without further degradation even after 600 shots. Such enhanced defects were indistinguishable from pre-existing defects and would otherwise have negligible effect on coating performance. We detected them only because we compared photographs of the sites both before and after irradiation. Some threshold morphologies yielded immediate massive damage necessitating the cessation of irradiation. This was particularly the case for some samples on Mo substrates or with Mo precoats and was also the case for most samples at higher-than-threshold fluences.

2.3. Task II experiments of HR coatings on dielectric and Mo substrates

The second series of experiments were based on the results of Task I as well as ancillary successful experiments conducted in support of the ICF program. The major thrust emphasized the following:

i. We concentrated on the more promising material combinations. These included both tantala/silica and two designs of hafnia/silica.

ii. We irradiated each site with ramped fluences thus conditioning the coating to produce higher damage thresholds.

iii. SPOC developed a composite substrate wherein a multi-layer dielectric overcoat was deposited on the Mo substrate and polished before the HR coating was deposited on it.

From previous work we had determined that the hafnia/silica material combination yielded among the highest and most consistent laser damage thresholds for dielectric HR coatings.[3] Moreover, we also found, particularly in the case of hafnia/silica HR's, that we could significantly and permanently increase the damage thresholds up to as high as a factor of three by laser conditioning or "annealing" the coatings with sub-threshold fluences.[4] We have investigated a variety of conditioning processes which can in fact be extended to large areas on full-sized optics such as are used in the Nova laser at LLNL.[5] In the current set of experiments we irradiated the samples in Task I with unconditioned fluences where we maintained nominally the same fluence for 600 shots at 10 Hz. We refer to these tests as S:1 meaning the "same fluence on one site" (Fig. 2). The tests for Task II were conducted with larger spot sizes (≥ 1.5-mm diameter, $1/e^2$) using ramped fluences. We call these tests R:1 meaning a "ramped increase in fluence on one site" (Fig. 2). This conditioned the samples at each site to the prescribed fluence levels. Fig. 2 shows that in the latter case, after reaching the desired fluence level, we then maintained the fluence fixed for the remainder of the irradiation so that each site was then irradiated at the final fluence with several hundred shots.

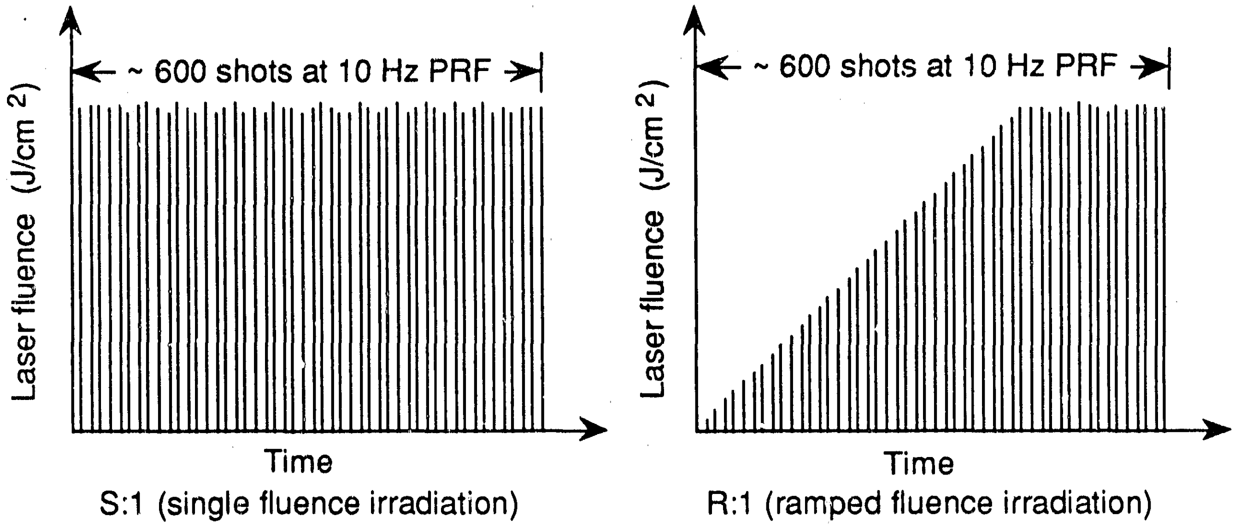

Fig. 2. Samples were typically irradiated with about 600 shots for one minute at 10 Hz PRF. With S:1 irradiation, the same nominal fluence was maintained for each shot. With R:1 irradiation we ramped up the fluence from near zero to the desired level and then maintained the same fluence for the remaining shots. The latter is a form of conditioning or "annealing" and can permanently raise the damage threshold.[4,5] S:1 irradiation was used for Task I; R:1 irradiation was used for Task II.

In order to attempt to resolve the problems caused by the residual large-sized defects on the Mo substrates, SPOC applied a variety of techniques for producing composite substrates. In all cases the substrates were coated with an optically thick material which could then be polished smooth prior to the deposition of the HR coating. Three processes were examined:

i. A thick layer of silica was sputtered on several substrates. None of these sample were, however, polished because they either crazed or delaminated due to high stress and poor adhesion.
ii. An alumina layer was flame-spray coated on another set of substrates. Although some regions of these substrates did have optically smooth surfaces, in general the polished surfaces were found to be too porous.
iii. SPOC deposited multiple thick layers of alumina and silica on several substrates. The coatings were stress balanced and designed for minimal optical effect at the HR design wavelength of 1064 nm.

In Fig. 3 we summarize the damage test results conducted on the samples from Task II. It should be noted that the fluence axis was compressed in order to reflect some damage thresholds almost twice as high as those depicted in Fig. 1. Besides the specific design differences in the coatings and composite substrates, these tests were conducted under the slightly different conditions described above. All of the Task II measurements were made with 8-ns pulses using ramped irradiation and with larger spot sizes, typically ≥ 1.5-mm diameter. From past experience we would expect higher thresholds because of the ramped conditioning and a nominal 10% lowering of thresholds because of the shorter pulse duration. We conducted some preliminary measurements with small spot sizes under both unconditioned and conditioned irradiations. We demonstrated an average improvement of 71% with conditioning. In addition, we also noted that the severity of damage at threshold was considerably less when the sites had been conditioned. The damage morphology was identical to that described for Task I.

Although the results for the Task II experiments were in general very encouraging they did raise a few questions. Some of the general conclusions from these tests were as follows:

i. For those coatings on dielectric substrates without Mo precoats we found that the thresholds ranked in the following order:

hafnia/silica (non-λ/4 layers)	46 - 71 J/cm^2
hafnia/silica (λ/4 layers)	30 - 57 J/cm^2
tantala/silica (λ/4 layers)	11 - 28 J/cm^2

Although the tantala/silica coatings were again cosmetically very clean looking they had notably lower thresholds than their companion pieces of Task I. This was in spite of the fact that they had a slightly lower density of pre-existing defects than the hafnia/silica coatings. We cannot account for this drop in thresholds.

ii. With but one exception these samples had an average 25% higher threshold when deposited on BK-7 as compared to fused silica. Cosmetically there was no difference between comparable coatings on the two different substrates.

iii. Other than for the same exception noted above, we found little difference in thresholds between coatings deposited in the large production chamber vs. the smaller production chamber.

iv. All of the coatings that were deposited on evaporated Mo precoats had relatively low thresholds ranging from 8 to 14 J/cm^2. However, we did note more pre-existing defects than were observed for such samples in Task I. Nevertheless, these thresholds were still comparable to or better than any previously tested enhanced or HR-overcoated metal mirrors.

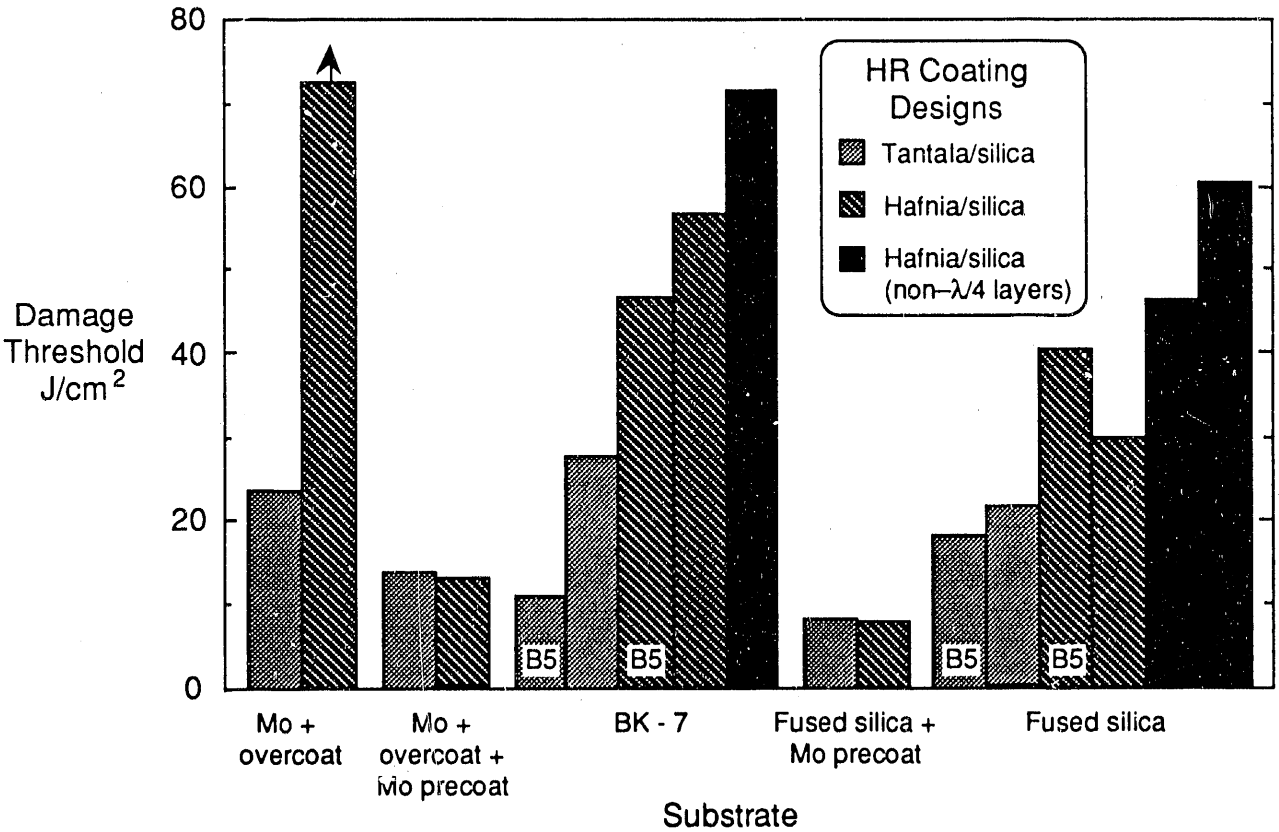

Fig. 3. Laser damage thresholds of three coating designs, on three substrate materials, with and without evaporated Mo precoats between the substrates and HR coatings (SPOC — Task II). The thresholds were measured at each site with 600 pulses ramped at 10-Hz PRF from zero to the desired test fluence (conditioning). We used Gaussian spot diameters ≥ 1.5 mm (1/e^2) at 1064 nm with 8-ns pulses. The arrow indicates that we were unable to damage the sample at the maximum test fluence shown. The samples labeled "B5" were coated in a small production chamber. The other samples were coated in a larger production chamber.

v. Probably the most noteworthy results were those for the first two samples in the chart. The coatings were deposited directly on the composite substrates consisting of polished alumina/silica overcoats on Mo substrates. The tantala/silica HR ranked well within the group of other tantala/silica coatings on dielectric substrates. We were, however, unable to induce even superficial damage on the hafnia/silica sample at any of 15 sites at fluences up to 73 J/cm². We did ultimately generate massive damage on one site at 85 J/cm² when we conducted small spot (0.7 mm) conditioning measurements with 10-ns pulses. Although the Mo substrate surface showed a typical high density of damage-prone defects, the polished overcoat was sufficiently smooth to allow a dielectric HR coating to be deposited uniformly. The damage threshold of this coating surpassed even those thresholds of coatings deposited on dielectric substrates.

3. HIGH PRF DAMAGE TESTS

All of theses samples were also subjected to damage tests on the Kilroy laser damage facility. This, as well as our other facilities and diagnostic systems, were described at the Boulder Damage Symposium in 1989.[6] The primary goal for this laser was to be able to conduct laser damage tests with rep-rated pulses at PRF's ranging up to 6 kHz at 1064 nm. Rather than multiplexing several lasers together in order to obtain such rep-rates, we chose to have fabricated a cw YAG "welder" laser with three heads in one laser cavity. We obtained the desired 6-kHz rep-rate by activating four separate acousto-optical Q-switches also distributed within the laser cavity. This generated temporally modulated pulses with an envelope of 65 ns FWHM. Because the laser produced a highly divergent, multi-mode beam of relatively low pulse energy, the highest fluences we were able to obtain ranged up to 10 J/cm² when focused to a spot size nominally 0.3 mm in diameter.

The first phase goal of this program was to establish whether dielectric HR coatings deposited on cooled metal substrates could survive FEL fluences as high as 2 J/cm² without succumbing to damage due to laser absorption at rep-rates up to 6 kHz. We irradiated the samples under a variety of conditions:

i. Each sample was irradiated at up to 10 separate sites with nominally 0.3-mm beams at fluences ranging from 7.3 to 10.0 J/cm². Each site was exposed to from 0.4 to 1.8 million shots.

ii. In addition, all samples in Task II were also irradiated with larger beams on the order of 0.5-mm diameter at fluences ranging from 2.4 to 3.7 J/cm² for 1.8 million shots.

iii. Several samples were scanned through the laser beam in 10-mm-long rows at rates of about 2 mm/minute thus subjecting a larger area of the coating to irradiation at fluences exceeding 8 J/cm².

We were unable to induce damage in any of the samples under these conditions. Moreover, for several samples we also monitored the temperature rise of the irradiated sites with an infrared camera. We measured barely perceptible temperature rises ranging up to 0.3° C for 5 minute exposures at the highest fluences. We were confident that we did indeed have laser fluences capable of generating observable damage under the right conditions. Several other coatings not related to this current study did damage at fluences ranging from < 1.7 to 9.0 J/cm². In addition, we observed damage on two samples in this work under extenuating circumstances. In one case we deliberately irradiated a site at 10 J/cm² that contained an obvious dust particle on the surface. In another situation we focused our laser beam down to a 1.5 mm spot size at an estimated fluence of 30 J/cm². In both cases the samples failed so catastrophically that we had to cease irradiation.

4. CONCLUSIONS

We have demonstrated that laser-conditioned hafnia/silica dielectric HR coatings can be fabricated to yield high damage thresholds exceeding 40 J/cm² at 1064 nm when irradiated with ramped fluences using 8- to 10-ns pulses. Comparable tantala/silica HR's performed less well. The failure mechanism in most of these coatings was the enhancement of isolated pre-existing artifacts usually ≤ 10 μm in size. These defects occasionally grew an insignificant amount but also formed the nuclei for larger delaminations and massive damage. These coatings have been found to be sufficiently uniform and free of pinholes to prevent laser light from penetrating to and damaging metal substrate surfaces at fluence ranging from 8 to 14 J/cm^2. Moreover, we have shown that this shielding of the metal substrate was significantly improved by first coating and polishing a dielectric overcoat on top of the substrate.

At high PRF's up to 6 kHz we have also demonstrated that such HR coatings on both dielectric and metal substrates survived small-spot fluences ranging up to 10 J/cm^2 with 65-ns pulses at 1064 nm. For spot sizes ranging from 0.3- to 0.5-mm diameter the absorption in the coatings caused only a negligible amount of heating. We were, however, limited by the available fluence of our laser to test such samples under larger laser-spot conditions. Beam areas one to two orders of magnitude larger at comparable fluences would be needed to fully establish the survivability to thermal loading of such coatings on full-sized laser optics.

Several of the issues raised in this work merit further study. The repeatability of the high performance of the hafnia/silica coatings needs to be established as well as the causes for inconsistent results with the tantala/silica HR's and the Mo precoats as manifested by the different results of Tasks I and II. Clearly, a major goal should be the further study of the beneficial effects of polished overcoats on substrates. The repeatability of high thresholds on Mo substrates as well as the promise of improved results of such a technique on dielectric substrates should be undertaken.

5. ACKNOWLEDGEMENTS

We wish to acknowledge the assistance of F. van Milligen and his colleagues at United Technologies Optical Systems, West Palm Beach, FL in supplying and polishing the molybdenum substrates which were used in the course of these experiments.

This work was performed jointly under the auspices of the US DOE by LLNL under W-7405-ENG-48 and for the DOD under SDIO/SDC MIPR No. W31RPD-0-D4074.

6. REFERENCES

1. F. Rainer, E. A. Hildum, and D. Milam, "Database of Average-Power Damage Thresholds at 1064 nm," Nat. Inst. Std. & Tech. (U. S.) Spec. Publ. 756, pp. 410-418, October 1987.

2. F. Rainer, R. P. Gonzales, and A. J. Morgan, "Laser Damage Database at 1064 nm," Nat. Inst. Std. & Tech. (U. S.) Spec. Publ. 801 (SPIE Vol. 1438), pp. 58-73, November 1989.

3. F. Rainer et al., "Damage Measurements on Optical Materials for Use in High-Peak-Power Lasers," Nat. Inst. Std. & Tech. (U. S.) Spec. Publ. 801 (SPIE Vol. 1438), pp. 74-83, November 1989.

4. C. R. Wolfe, M. R. Kozlowski, J. H. Campbell, F. Rainer, A. J. Morgan, and R. P. Gonzales, "Laser Conditioning of Optical Thin Films," Nat. Inst. Std. & Tech. (U. S.) Spec. Publ. 801 (SPIE Vol. 1438), pp. 360-375, November 1989.

5. M. R. Kozlowski, C. R. Wolfe, M. C. Staggs, and J. H. Campbell, "Large Area Laser Conditioning of Dielectric Thin Film Mirrors," Nat. Inst. Std. & Tech. (U. S.) Spec. Publ. 801 (SPIE Vol. 1438), pp. 376-390, November 1989.

6. A. J. Morgan, F. Rainer, F. P. Morgan, R. P. Gonzales, M. R. Kozlowski, and M. C. Staggs, "Expanded Damage Test Facilities at LLNL," Nat. Inst. Std. & Tech. (U. S.) Spec. Publ. 801 (SPIE Vol. 1438), pp. 47-57, November 1989.

- END -

DATE FILMED

01 / 29 / 91