A Study of Laser Conditioning Methods of Hafnia Silica Multilayer Mirrors

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A study of laser conditioning methods of hafnia silica multilayer mirrors

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ABSTRACT

A variety of 1064-nm laser conditioning methods were investigated to find the optimum method for production of large aperture (0.25 m²) multilayer coatings for the National Ignition Facility and Laser MegaJoule. Two conditioning methods were explored, multi-step and single-step on two different laser systems. Off-line conditioning was done on PLATO, a small beam diameter (≈1 mm) raster scanning system. On-line conditioning was done on Beamlet, a single beam prototype of the National Ignition Facility with a large rectangular beam (35 cm × 35 cm). Concurrent with this work, coating process development for low-defect density high damage threshold coatings was realized by switching from hafnia to hafnium starting materials. The results of this study suggest that single-step raster-scan off-line laser conditioning is an effective method to raise the damage threshold of multilayer mirrors deposited from hafnium and silica by reactive electron beam deposition.

Key words: laser conditioning, laser-induced damage, laser damage morphology, hafnia-silica multilayer coating

1. INTRODUCTION

Large aperture fusion lasers operate near optic damage thresholds to cost-effectively maximize system performance. Therefore methods to increase the damage threshold of 1064 nm fluence-limiting multilayer optical coatings are under investigation. Previous work has demonstrated that large-aperture laser conditioning is a viable technology for increasing laser damage threshold. However, over the course of a three year production phase approximately 1,100 mirrors and 200 polarizers will require laser conditioning for the National Ignition Facility (NIF) so the process must be optimized for a production operation.

The laser conditioning process consists of irradiating a coating by starting at a low fluence and gently ramping to the system operation fluence. The most widely understood conditioning mechanism is nodular ejection, although other mechanisms may be present. In an effort to reduce the number of fluence limiting defects, coating development efforts have concentrated on source stabilization, particularly for the high index material since it is the dominant defect source. Source stabilization has been accomplished by replacing the starting material from an oxide to metallic state of hafnium. In parallel, a developmental effort is ongoing to minimize the laser conditioning time by reducing the number of conditioning steps.

Nodular ejection is laser damage because it is a surface modification. However, if the conditioning-induced damage does not grow with subsequent irradiation, it will not limit the optic lifetime on NIF. Of equal importance is whether the damage impacts the NIF performance. The criteria used for NIF is that the damage size does not exceed 280 μm in diameter and the minimum obscured area does not reduce the components average reflectance below specification.

2. EXPERIMENTAL PROCEDURE

Two facilities are available at Lawrence Livermore National Laboratory (LLNL) for studying large area conditioning phenomena including a small-beam raster-scan instrument termed Probed Large Area Testing of Optics (PLATO) and Beamlet, a prototype laser constructed to validate performance expectations of a single NIF beamline. These systems are used to investigate laser conditioning under a small set of variables including small beam versus large beam, hafnia versus hafnium, and single-step versus multi-step.
The experimental set up is described elsewhere.12 Twelve mirrors with dimensions of 40 cm × 57 cm were deposited by reactive electron-beam deposition from hafnia or hafnium and silica by four different coating vendors. These mirrors were divided into three different experiments as defined in table 1. The experiments on Beamlet, with a 30 cm × 30 cm beam, were configured as shown in figures 1a and 1b. On typical damage test systems, the diagnostic wedge is located before the test optics for characterization of the beam before striking the optic. However, in this setup the mirrors were placed in a mirror tower upstream of the diagnostic beamsplitter. This configuration was selected because of the difficulty and cost associated with repositioning the diagnostic optics for controlling the 10 input power and wavefront of the beam for future KDP frequency conversion studies.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Coating vendor</th>
<th>Starting material</th>
<th>Beam size (in mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Role of conditioning beam size</td>
<td>A &amp; B</td>
<td>HfO₂ &amp; SiO₂</td>
<td>1 (diameter) &amp; 300 (square)</td>
</tr>
<tr>
<td>Role of starting material composition</td>
<td>A</td>
<td>HfO₂ or Hf &amp; SiO₂</td>
<td>1 (diameter)</td>
</tr>
<tr>
<td>Influence of number of conditioning scans</td>
<td>A, B, C, &amp; D</td>
<td>HfO₂ or Hf &amp; SiO₂</td>
<td>1 (diameter) &amp; 300 (square)</td>
</tr>
</tbody>
</table>

Fig. 1a Depiction of mirror tower placement in Beamlet relative to diagnostic systems.  
Fig. 1b Depiction of mirror locations within the Beamlet mirror tower.

The PLATO system, used for small beam conditioning studies, consists of a stage that raster scans an optic past a stationary 1 mm diameter beam as illustrated in figure 2.12 A number of in-situ diagnostic systems measure the beam diameter, beam energy, and scatter from a HeNe probe beam of each beam position before and after irradiation. This information is used to study the areas with the largest change in scatter for damage size determination by optical microscopy.

2.1. BEAM SIZE (OFF-LINE VERSUS ON-LINE LASER CONDITIONING)

There are a number of significant differences between laser conditioning with small-beam off-line raster-scanning and large-area on-line fixed position. Small-beam systems can be installed at coating vendors for post-processing coatings to increase their damage threshold. They also serve as quality assurance tools to verify process stability and compliance with the full-aperture laser damage threshold specification. Unfortunately small-area damage tests on small optics have had little correlation with the laser damage thresholds of low-density fluence-limiting defects on these large optics. With current technology, a single raster scan of a typical NIF optic takes approximately twelve hours.

The optics could also be conditioned during the activation of NIF with a full-aperture beam to eliminate the costs associated with remote small-beam conditioning systems. Since the activation of each NIF beam will likely consist of a fluence ramp starting at a low fluence, no additional conditioning costs would be incurred during NIF activation. However, during operations, damaged optics replaced with unconditioned spares will require a conditioning fluence ramp that would likely impact the

Fig. 2 Depiction of PLATO laser conditioning process.
experimental schedule. The time to laser condition an optic with a large beam is limited by the NIF repetition rate of one shot every eight hours.

For both of these conditioning techniques, the beam shape has a significant impact on the fluence uniformity across the entire clear aperture. The small-beam packing density impacts the spatial uniformity of the fluence. The current method, used for the initial set of Beamlet optics, has a beam increment between pulses equal to the beam diameter at 90% of the peak intensity as illustrated in figure 2. With no beam pointing instabilities, the packing density would be as illustrated in the beam diameter at 90% of the peak intensity, obviously improvements in beam centration stability would result in improved fluence uniformity across the coated optic. Given a nearly perfect Gaussian beam, the minimum fluence for the two extreme centration cases can be calculated as follows:

\[
I \left( \sqrt{2} r \right) = I_o \exp \left( \frac{-2r^2}{\omega_o^2} \right) = I_o \exp \left[ \frac{-r^2}{\omega_o^2} \right]^2 = I_o (0.9)^2 = 0.81 I_o \\
I \left( (2 + \sqrt{2}) r \right) = I_o \exp \left( \frac{-2 + \sqrt{2} r^2}{\omega_o^2} \right) - I_o \exp \left( \frac{-r^2}{\omega_o^2} \right)^2 = I_o (0.9) (2 + \sqrt{2})^2 = 0.29 I_o
\]

Minimum intensity for optimum beam pointing.

Minimum intensity for worst case beam pointing.

Large-area conditioning has a similar packing density issue because the NIF beam has a modeled intensity modulation of 2:1 at the transport mirror planes. The modulation has both systematic features from damaged and contaminated upstream optics and random features. The random nature of the spatial position of the peak fluence is problematic since the peak fluence could easily align to an area that is conditioned to only the average fluence (50% peak) or lower resulting in a high probability of damage. The NIF transport mirrors are used for beam pointing and centering to the target chamber, hence some beam wander across the optic surface may occur between shots due to thermal drift. As illustrated in figure 4, the clear aperture is larger than the conditioning beam area, therefore an unconditioned area will exist after on-line conditioning. This unconditioned area could be exposed to a realigned high fluence shot resulting in a high probability of laser damage to the coating. This issue could be solved by a conditioning routine with the beam positioned at each corner of the optic for complete coverage. However, this would be extremely difficult to accomplish on-line during NIF activation so a separate beam line would have to be dedicated to large area laser conditioning.

2.2. STARTING MATERIAL COMPOSITION

The starting material composition for the hafnia layers has a direct impact on the concentration of defects imbedded in the coating. The current proposed defect ejection mechanisms in hafnia are expansion-induced internal stresses as a result of a crystalline phase transformation within the material charge and e-beam exposure of internal voids as a result of poor packing density. Hafnium charges are typically denser and there is no temperature induced phase transformation, hence a 3-10x defect density reduction is realized by evaporating hafnium. A similar reduction in the number of visible plasmas is observed during
the laser conditioning process. Previous defect studies of hafnia and silica deposited coatings revealed almost exclusively hafnia seeds. Interestingly, coatings deposited from hafnium have defect-seed cross-sections of predominately silica seeds at a significantly lower density. Therefore it is believed that defect seeds are generated from both starting materials, but the defect flux deposited from each source material composition is significantly different.

2.3. NUMBER OF CONDITIONING STEPS

The number of required laser conditioning steps is not well understood. The current theory is that damage is minimized by ejecting a nodule close to its ejection fluence. Also partial ejections require multiple shots to completely remove the nodular debris. To minimize damage, this cleaning process should likely be done at as low a fluence as possible. Therefore it is desirable to have a very shallow fluence ramp with many conditioning steps. Unfortunately the time constraints of twelve hours for small-beam and eight hours for large-beam conditioning are prohibitive for a large number of conditioning steps in a production environment.

A six-step raster-scan small-beam routine was used for the initial high fluence Beamlet optics. The number of scans was determined by small-area tests that demonstrated no noticeable difference in the damage threshold of a coating using the standard R:1 conditioning process versus a six-step approach. The standard R:1 test consists of 600 pulses over 1 minute at a repetition rate of 10 Hz with a fluence ramp during the first half of the test and a constant peak fluence during the second half of the test. The fluence of the first scan was selected to be one half of the unconditioned damage threshold as measured on a 50 mm diameter damage test witness coated in the same run. The fluence ramp was equally spaced to the NIF operating fluence as illustrated in figure 4a.

By exploiting the Gaussian shape of the conditioning beam, a fluence ramp may be realized with a single scan at the NIF operating fluence as illustrated in figure 4b. With this technique the fluence nonuniformity due to the beam wander becomes a more significant issue since multiple scans allow some averaging to smooth out the fluence spatial distribution. This technique is only relevant for the small beam raster-scanning systems because the large-aperture NIF beam is approximately flat top. Single-step conditioning is significantly less costly due to the reduced number of required conditioning stations and fewer operating hours.

![Fig. 4a Depiction of six-step laser conditioning process.](image)

![Fig. 4b Depiction of single-step laser conditioning process.](image)

3. RESULTS

Three experimental campaigns were conducted on Beamlet as part of the laser conditioning development program. The experiments were designed for validation of the technology without damaging the optics because of the experimental cost associated with manufacturing the optics and Beamlet operations and the eventual requirement to realign the Beamlet laser with the mirror tower into the diagnostic systems. This approach is well suited for studying optic lifetimes, but not for determining the safety margin of the coating's laser-conditioned damage threshold over the peak operating fluence. Although unplanned, the first two experiments did offer an opportunity to damage the optics due to high modulation in the Beamlet laser most likely caused by plasma-induced pinhole closure. Unfortunately camera saturation and the diagnostic splitter location negated the ability to calculate the peak fluence, but minimum damaging fluences could be determined.
3.1. BEAM SIZE (OFF-LINE VERSUS ON-LINE LASER CONDITIONING)

The first experiment was designed to determine if there was any performance differences when laser conditioning with different beam sizes. Two different vendors, both using hafnia and silica as starting materials coated the mirrors. One mirror from each vendor was laser conditioned using a six-step small-beam raster scan process to 32 J/cm² at a 9 ns pulse length. This correlates to 22 J/cm² for a 3 ns pulse length when using a $t^{0.35}$ scaling relation. These mirrors were placed in the upper (downturn) locations at positions LM7 and LM8. An unconditioned mirror from each vendor were placed in the lower (upturn) locations at positions LM6 and LM9 on a rail system for ease of removal. In addition, the lower mirrors could be translated to an unconditioned area thus simulating an on-line conditioned NIF optic that is irradiated in an unconditioned area due to pointing and centering tolerances. The mirrors are enclosed in a clean sealed environment to minimize contamination of the coated surfaces.

The shot sequence is illustrated in figure 5 for the two campaigns. In both cases, Beamlet was configured to yield a 3-ns square pulse. As stated earlier, excessive modulation of the beam was observed which lead to damage of some of the mirrors. During the first campaign, only the on-line conditioned mirrors damaged after four shots indicating only partial conditioning. This result also validates that laser conditioning is required to meet the NIF operating fluence for the current coating technology. Scatter maps of the LM6 mirror after damage indicate a strong correlation between high scattering damaged areas and fluences exceeding 15 J/cm² as illustrated in figure 6.

![Fig. 5 Comparison of fluence ramps for off-line and on-line laser conditioning.](image)

Both campaigns were terminated due to severe modulation of the laser beam resulting in high peak fluences.

The second campaign consisted of the full six-step conditioning routine, although the fluence of the first shot was repeated twice due to the inability to precisely set the Beamlet output energy. The campaign was terminated at the last conditioning step because of laser damage from excessive intensity modulations in the beam. Again the on-line conditioned mirrors damaged, but so did one of the off-line conditioned mirrors indicating that the LM6 and LM9 mirrors were nearly fully conditioned. A comparison of LM6 mirror post-damage scatter maps indicates a strong correlation between high-scattering damaged areas and fluences exceeding 30 J/cm² as illustrated in figure 6. The difference in the damage morphologies shown in figures 6c and 7c is attributed to manufacturing process differences at the two vendors resulting in different degrees of film to substrate adherence and coating material properties.

3.2. STARTING MATERIAL COMPOSITION

Multilayers deposited from hafnia have a 3-10x reduction in defect density with fewer plasmas created during the laser conditioning process. The magnitude of the defect reduction varies with coating vendor based on their understanding and ability to control the process variables that contribute to source ejection. The damage thresholds on 50-mm diameter witness samples were comparable, independent of starting material composition, for coatings with less than 100-ppm absorbance. Because of the required oxidation, film stoichiometry was a major concern in the initial development of the hafnium process. Low absorbing films were created by process optimization with emphasis on appropriate oxygen flow control.

Three of the four coating vendors elected to coat their mirrors from hafnium for the third and final Beamlet campaign because of the reduction in laser conditioning-induced damage sites and no significant damage threshold difference between hafnia deposited coatings. A significant amount of plasma scalding was present on the hafnia deposited mirror, but not on the remaining three hafnium deposited mirrors. The plasma scalding is most likely due to variances in the coating deposition
process other than starting material composition because hafnia deposited mirrors coated by other vendors also had significantly less plasma scalding.

In figure 8 a significant reduction in scatter after small-area raster-scan laser conditioning is observed, indicating fewer conditioning-induced laser damage sites on mirrors coated by the same vendor, but from hafnium instead of hafnia starting materials. It should be noted that the number of conditioning steps for the hafnia deposited mirror was six, whereas it was only one for the hafnium deposited coating. However, a significant scatter change was observed after each conditioning step of the hafnia deposited mirror. Although this represents the extreme range because other hafnia deposited mirrors had less change in scatter; the hafnium deposited mirrors always had less laser conditioning-induced damage. Due to differences in the scatter diagnostic hardware and scatter dependence on the mirror pass-band reflectance, observable differences in the background scatter of the images of the two different mirrors is not significant.

In this third campaign, no significant modulation was present as illustrated in figure 9 to damage the coatings, therefore it was impossible to assess whether the hafnium deposited mirrors truly required laser conditioning. However the full-aperture quality assurance capability alone justifies continuing to laser condition hafnium deposited mirrors.
One of the mirrors developed a damage site after the first NIF Haan Pulse. The NIF Haan pulse is a nominally 20-ns long pulse with the last 3 ns at significantly higher fluence than the first 17 ns. The damage did not correlate to any existing beam modulation intensity peaks or notable conditioning-induced damage. However, the mirror was placed in an upturn location and dust was clearly visible on the coated surface. It is likely that the cause of the damage was contamination, but other causes have not been eliminated. After replacement with the only mirror that survived the first two Beamlet campaigns (also coated by the same vendor), the remaining three hafnium deposited mirrors were in service on Beamlet for approximately nine months until the Beamlet laser was decommissioned.

Fig. 8 Scatter maps of optics deposited from either hafnia or hafnium before and after laser conditioning.

Fig. 9 Fluence ramp for the third and final Beamlet campaign to validate performance of single-step laser conditioned coatings deposited from Hafnium.
3.3. NUMBER OF CONDITIONING STEPS

During these campaigns, both six-step and single-step conditioned mirrors were successfully laser conditioned. In addition to the initial four single-step-conditioned mirrors, one mirror from two different vendors had a few 1-mm diameter damages after single-step conditioning. These damage sites grew when irradiated at the NIF average fluence, hence they would limit the optic lifetime. At this time it is unknown if the laser damage is a result of the more aggressive single-step laser conditioning process or the use of hafnium as the starting material.

4. ANALYSIS

The damage threshold of optical thin films is dictated by low-density fluence-limiting defects. Therefore full-aperture damage thresholds must be determined by irradiating the entire surface. The process of raster scanning a small Gaussian beam across the entire surface is adequate for determining the damage threshold, but not for establishing the effectiveness of laser conditioning because the coating is being single-step conditioned. The severely modulated large-area Beamlet beams provided a unique opportunity to determine both the damage threshold of a conditioned coating over reasonably large areas and the damage threshold improvement by laser conditioning of coatings deposited from hafnia and silica.

There was no excessive intensity modulation during the final campaign with the mirrors deposited from hafnium and silica, so the damage threshold and laser conditioning damage threshold improvement was not determined. Alternatively, the lack of an available top hat small-aperture beam and the poor packing density needed to prevent conditioning by increasing the spacing between small-aperture Gaussian beams, also prevented the determination of the full-aperture conditioning effect on coatings deposited from hafnium and silica. Although there is a significantly lower defect density in hafnium deposited coatings; the presence of plasmas indicates nodular ejections. However, laser conditioning occurs only if nodules are ejected below the operating fluence, a pit remains that does not change at the operating fluence, and the pit would have grown if ejected at the operating fluence. Although unlikely, the defect reduction caused by the change in starting material composition may have eliminated the class of defects that laser condition.

A potentially useful method of studying laser conditioning is by photothermal microscopy. Current work on small samples with photothermal microscopy shows a correlation between defects with high photothermal signal and low damage threshold. This tool provides a nondestructive means of locating the fluence limiting defects to study effective laser conditioning methods. Unfortunately the current technology lends itself to only small sample sizes due to the amount of time required to scan the sample.

There is a statistical nature to the fluence requirements of the NIF mirrors. The mirrors need to survive a limited number of shots dictated by a shot cycle of three shots per day over six days per week resulting in around 900 shots per year. Mirrors from NOVA, the current generation fusion laser, have survived as long as fifteen years without replacement, but at lower fluence. Only a small percentage of the NIF shots are expected to be at the peak NIF fluence since not all experiments are conducted at high fluence. Finally, only a small percentage of the beam is at the peak fluence. Therefore mirrors with fluence limiting defects have a probabilistic lifetime determined by the requirement for alignment of fluence limiting defects with the peak fluence locations of the beam that exceed the growth threshold of the damage site.

Finally, contamination may have played a role in the damage of the single-step laser conditioned mirror deposited from hafnia and silica. The mirrors on Beamlet are in a sealed enclosure with no active filtration. The upturn mirrors required periodic cleaning by blowing deionized Nitrogen across the surface to remove particulates. Because it was difficult to remove the downturn mirrors, the two mirrors with the most conditioning-induced laser damage were put in the upturn positions for ease of removal in the event of laser damage. Therefore it is difficult to determine the significance of the contamination effect.

5. SUMMARY

One-step laser conditioned mirrors deposited from hafnium and silica at multiple coating vendors were successfully deployed for routine operations on Beamlet, a single beam prototype of NIF. Therefore a cost-effective production-compatible process is available for NIF optics fabrication, although some further work is required to determine a slightly gentler laser conditioning routine to improve production yields and optic lifetime. The mirrors deposited from hafnia have a large-area damage threshold of 30 J/cm² as demonstrated on Beamlet. The large-area damage threshold for hafnium deposited mirrors is
unknown since no significant modulation was observed during Beamlet tests, but the threshold does exceed the peak raster-scanned fluence of 22 J/cm². Additionally, it is not proven that laser conditioning is required for mirrors deposited from hafnium, but the presence of low-density fluence-limiting defects necessitate full-aperture survivability scans.

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7. REFERENCES


