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The advantages of evaporation of Hafnium in a reactive environment to manufacture high damage threshold multilayer coatings by electron-beam deposition

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ABSTRACT

Electron-beam deposition is the current method to produce large-aperture high laser-induced damage threshold coatings for the National Ignition Facility, a 1.8 MJ fusion laser. The e-beam process is scalable to large optics up to 0.25 m^2 and with laser conditioning has relatively benign coating defect ejections resulting in high damage threshold thin films. The latest technological breakthrough in manufacturing high damage threshold coatings is e-beam deposition of hafnia by evaporation from a metallic instead of an oxide source in a reactive environment. Although the damage threshold is not significantly increased, a 3-10× defect reduction occurs resulting in significantly less coating modification during laser conditioning. Additional benefits of this technology include improved interfaces for the elimination of flat-bottom pits and up to 3× reduction in plume instability for improved layer thickness control and spectral performance.

Key words: Hafnium, hafnia, electron-beam deposition, laser-induced damage, and multilayer coating

1. INTRODUCTION

One of the challenges behind constructing the National Ignition Facility (NIF)[1], a 1.8 MJ, 192-beam Inertial Confinement Fusion (ICF) laser facility, is the production of large precision optics that are resistant to high laser fluence. The NIF laser will be used to support the nuclear stockpile stewardship program, study fusion reactions for a potential future energy source, and increase our understanding of cosmological events. The 1053-nm, 3-ns peak operating fluence of the NIF laser is 11 and 22 J/cm^2 on the polarizers and transport mirrors respectively. This is a 4× increase over the fluences used on NOVA, the current fusion laser at Lawrence Livermore National Laboratory (LLNL).

In 1995, a four-year coating development program was initiated to improve the quality of optical thin films for high fluence use in this next-generation ICF laser. The emphasis of this program was to increase layer thickness control for improved spectral performance and greater manufacturing yields, reduce coating stress to minimize laser beam wavefront distortion, and improve laser damage resistance to maximize component lifetime. Although much of the development program focussed on construction of vendor proprietary instrumentation, some of the process improvements can be reported.

As a result of this development program, two significant process modifications were adopted that directly involve coating materials. The oxygen partial pressure is now tuned during silica evaporation for low stress as well as low absorption coatings for either a low or ambient humidity environment depending on the installation location within the NIF laser.[2] A more dramatic process material change is the use of Hafnium instead of hafnia as the starting material evaporated in a reactive environment to produce hafnia films.[3] The advantages of evaporation from Hafnium are a more stable deposition process and higher quality thin films. The emphasis of this paper will be on the impacts of this starting material compositional change on optical coating properties and performance.

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2. DEFECT REDUCTION

Prior to embarking on the NIF coating development program, a significant LLNL study focussed on coating defect characterization.[4-5] By using an Atomic Force Microscope (AFM) and laser damage testing, it was found that nodular defects were determined to be the dominant coating defect that limited the laser fluence resistance of hafnia silica electron-beam deposited multilayer coatings.[6-7] Through the use of semiconductor circuit repair technology, a Focussed Ion Beam (FIB) was used to cross section and image nodular defects. By comparing the image contrast of nodular defect cross section micrographs from a Scanning Electron Microscope (SEM) and FIB, it was inferred that the majority of the seeds were caused by hafnia source ejections as illustrated in figure 1a.[8] The edges of the hafnia seeds were either rough or smooth indicating solid or molten source ejections, respectively.

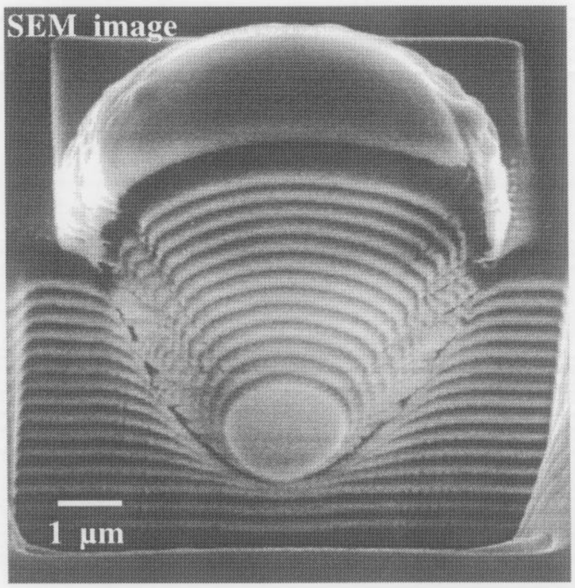


Fig. 1a Focussed ion beam cross section of a nodular defect caused by a molten hafnia seed.

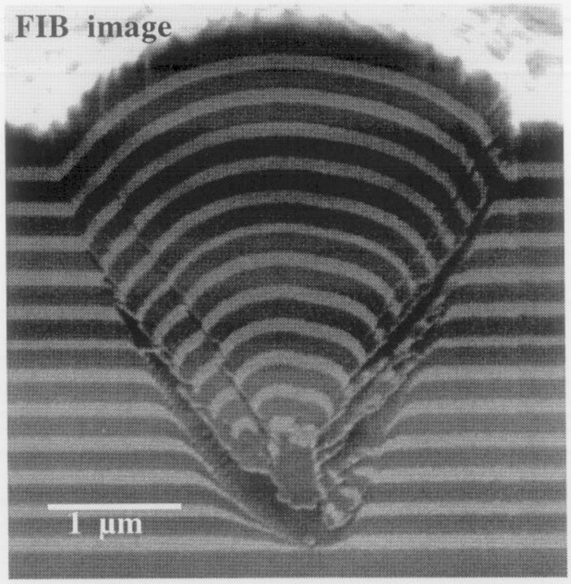


Fig. 1b Focussed ion beam cross section of a nodular defect caused by a solid silica seed.

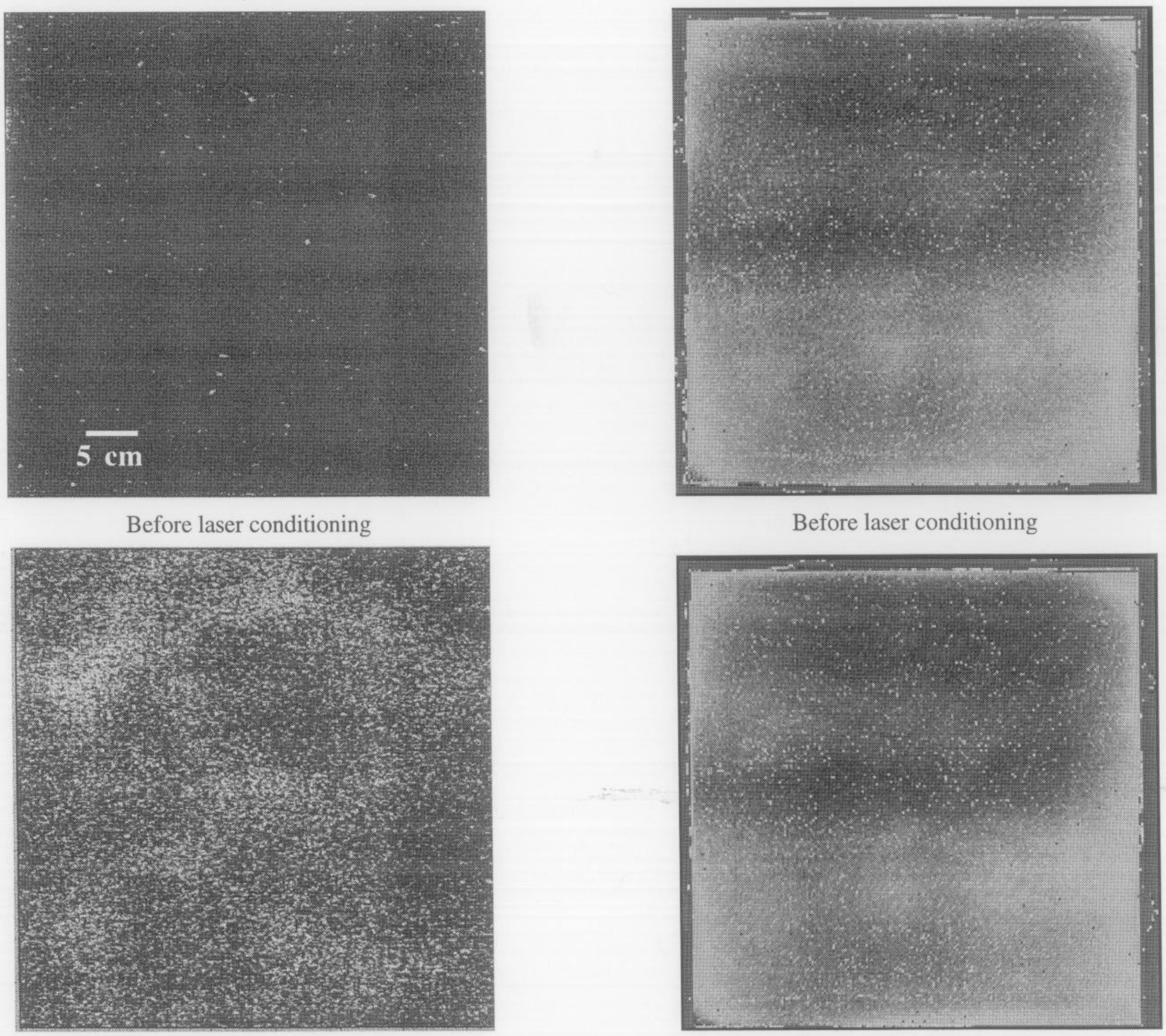
In an effort to reduce the density of coating defects, evaporation from Hafnium in a reactive environment was attempted. Because Hafnium has a higher thermal conductivity than hafnia, denser plugs of starting material can be created thus reducing the probability of exposing a void in the plug with an electron beam resulting in source spatter. Also Hafnium has no temperature-induced phase transformations between the cooled edges of the Hafnium plug in contact with the water-cooled crucible and the molten surface in contact with the electron beam. Hafnia, however, has a monoclinic to tetragonal phase transformation resulting in a 3.8% volumetric expansion inside the plug creating stresses that could result in particle ejections.[3] After switching to Hafnium starting material, the defect density dropped by a factor of 3 to 10 with the majority of coating defects now of silica composition as illustrated in figure 1b.[9]

Because the complete removal of all coating defects appeared to be a such a daunting task, laser interaction studies were instituted with an AFM to understand which defects should be eliminated. Or alternatively, how to minimize the damage caused by ejection of these nodular defects under high fluence laser irradiation. Although variances occurred in coatings deposited by different coating vendors, it was found that nodules under 0.5 – 0.7 µm in height did not damage at fluences greater than the NIF requirement.[7] It was also determined that as the defect height increased, the probability of damage increased. To minimize this damage, laser conditioning by irradiating defects with a gentle fluence ramp, proved to be an adequate means to increase the damage threshold by greater than 2× to meet the NIF fluence requirement.[9-11] Although the complete laser conditioning mechanism is not well understood, this process does reduce the probability of catastrophic nodular ejections and instead creates benign pits that tolerate much higher fluences.[12]

The impact of changing to a metallic form of starting material also affected the laser conditioning process.[9] For a Hafnium deposited coating, a 3-10× reduction in the number of plasmas created during laser conditioning was observed over hafnia deposited coatings. Plasmas are typically created during nodular ejection so a reduction in plasma occurrences indicates fewer nodular ejections. This observation is consistent with the defect reduction that occur in Hafnium deposited coatings. It also indicates that some conditionable defects still remain in Hafnium deposited coatings.

The laser conditioning system at LLNL has a 633 nm scatter diagnostic to monitor the coating before and after laser conditioning. For high reflector coatings laser conditioned at 1064 nm, a significant reduction in the number of scatter sites occurs for Hafnium deposited coatings as illustrated in figure 2. The scatter diagnostic was upgraded between conditioning of the two mirrors so only comparisons of the differences before and after conditioning are meaningful. Despite the scatter observed in the hafnia film at 633 nm, both the hafnia and the Hafnium deposited films meet the NIF reflectance requirement of greater than 0.995 at 1053 nm, however, the lower level of surface modification after laser conditioning seen in the Hafnium derived films is preferred.

Beamlet mirror deposited from hafnia | Beamlet mirror deposited from Hafnium



After laser conditioning at 22 J/cm^2 | After laser conditioning at 22 J/cm^2

Fig. 2 633 nm scatter maps of optics deposited from hafnia or Hafnium before and after 1064 nm laser conditioning.

3. INTERFACE IMPROVEMENT

Another damage morphology typical of NIF coatings are flat-bottom pits as illustrated in figure 3.[13] This interfacial damage generally occurs at the multilayer interfaces that align to the standing-wave electric field peaks within a multilayer high reflector stack. The pit depth is typically 2, 4, or 6 layers deep corresponding to the first three electric field peaks at multilayer interfaces. In a quarter-wave high reflector design, these are the hafnia over silica interfaces. Hafnium deposited coatings with electric field peaks shifted to both interfaces also show preferential interfacial damage at the hafnia over silica interface.[14] It has been proposed that flat-bottom pits are created by heating of nanoscale absorbing seeds with fractures propagating along weaker interfacial boundaries as indicated by small divots or hillocks centrally located within the flat-bottom pit.[15]

Although flat-bottom pits occur to coatings deposited from hafnia, they did not normally occur to coatings deposited from Hafnium unless the interfacial electric fields are abnormally high. TEM cross sections of multilayer films in figure 4 prepared from both hafnia and Hafnium starting materials illustrate significant differences in the silica over hafnia interfacial quality due to the difference in number of interfacial voids. Although this is not the interface where flat-bottom pits typically occur, it is possible that the hafnia over silica interface quality is also improved for films deposited from Hafnium. It is also unknown if the starting material composition has some affect on the presence or absence of nanoscale absorbing seeds.

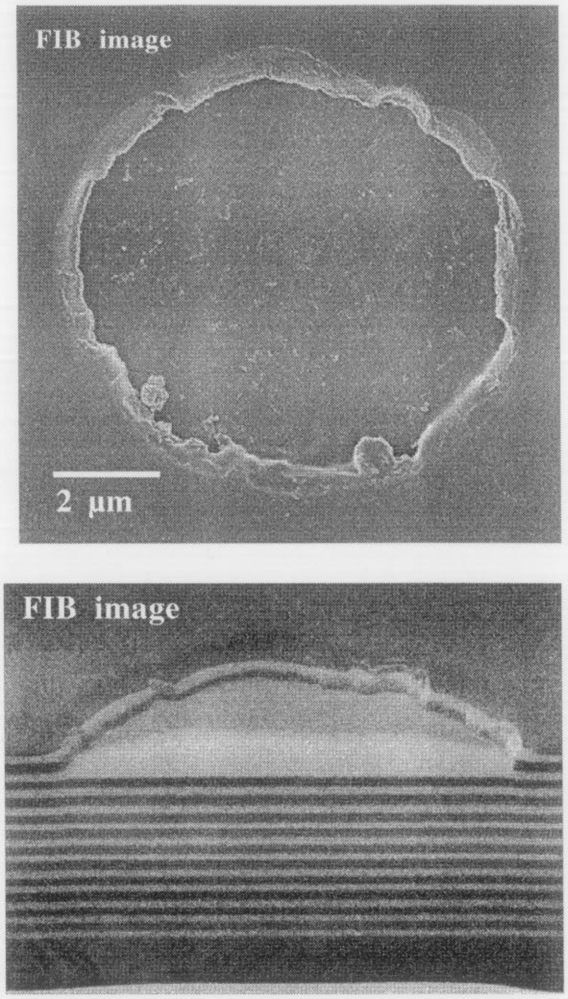


Fig.3 Top and cross sectional view of a four-layer deep flat-bottom pit damage site.

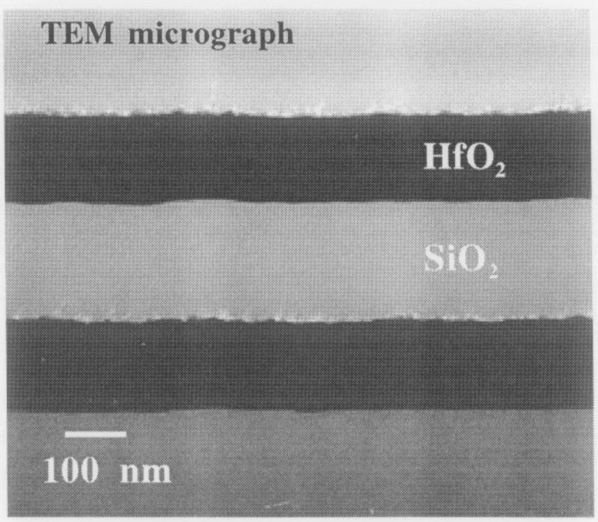


Fig 4a Multilayer film evaporated from hafnia starting material. Note voids in the silica over hafnia interface.

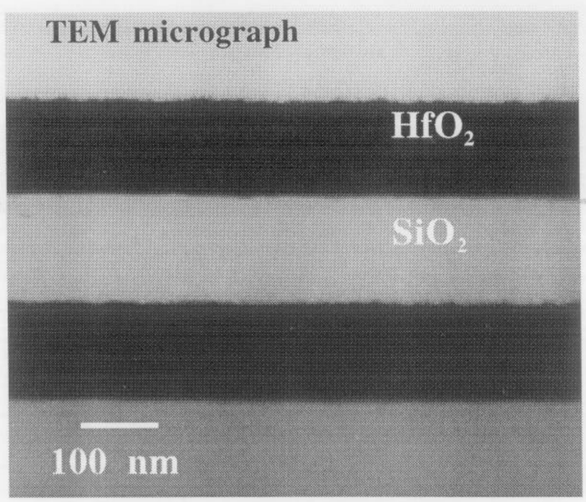


Fig. 4b Multilayer from Hafnium. Note absence of voids in silica over hafnia interface.

4. DEPOSITION STABILIZATION

One of the approaches in the NIF coating development program to achieve improved spectral performance and less defect generation was though stabilization of the deposition process. The traditional approach to hafnia evaporation has a few inherent stability problems. Hafnia starting material comes in a variety of forms including pellets, irregular chunks, and pill shapes. Each form must be premelted into a crucible to generate a plug of sufficient volume of material for deposition of multilayer optical thin films. Since the thermal conductivity of an oxide is low, there are typically gas voids entrapped within the material charge where incomplete melting occurred between pieces of starting material. These gas voids, once exposed to the electron beam can expel particulates and generate an unstable deposition plume.

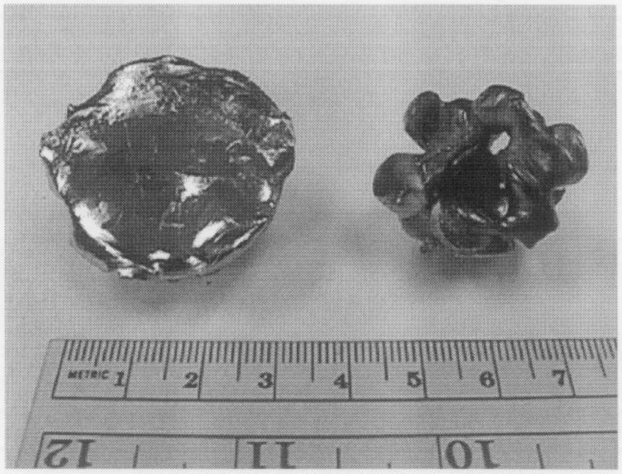


Fig. 5 Hafnium (left) and hafnia (right) melts after use in an e-beam crucible. The Hafnium forms a smooth pool while the high degree of topography in the hafnia melts results in vapor plume instability.

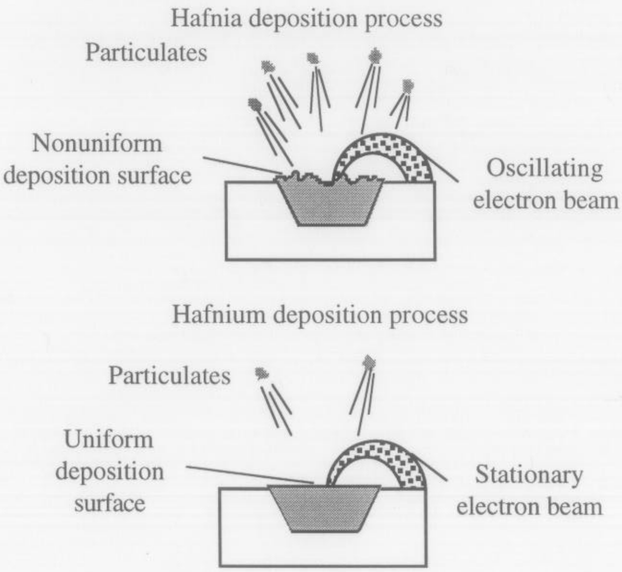


Fig. 6 Source material composition impacts e-beam interaction with the source material and density of particulate formation.

The top surface of the hafnia melt is typically very irregular as illustrated in figure 5. Since the thermal conductivity of hafnia is low, the electron beam must be swept across the surface to prevent the beam from drilling a hole in the surface of the material. Once the electron beam begins to drill a hole, the vapor plume is greatly reduced which changes the monitor-to-work ratio and coating uniformity. Conversely, the action of sweeping the beam over the irregular surface produces a very unstable and random vapor plume that also contributes to uncertainty in layer thickness.

In contrast, Hafnium has a much higher thermal conductivity, therefore even though the starting material needs to be melted to generate a solid plug, the probability of gas voids are significantly lower. Also the higher thermal conductivity of the Hafnium material allows for a stationary electron beam without drilling into the material as illustrated in figure 6. Deposition plume studies as measured with multiple quartz crystal monitors in coating chambers dedicated to ICF coatings, have revealed a 3× reduction in the plume instability after switching from a hafnia to Hafnium starting material.

Although the plume is significantly more stable, the ultimate requirement is to improve layer thickness control to manufacture designs with near theoretical performance. As an example of the spectral improvements realized during the NIF coating development program, comparisons can be made on hafnia deposited coatings manufactured for the Beamlet laser,[16] a single beam prototype of the NIF laser, and early Hafnium deposited coatings. Both of these coatings were manufactured without the aid of the improved spectral monitoring equipment. The polarizing bandwidth of the hafnia deposited coatings is only 50% of the theoretical bandwidth; whereas the Hafnium deposited coating is 80% of the theoretical bandwidth as illustrated in figure 7.

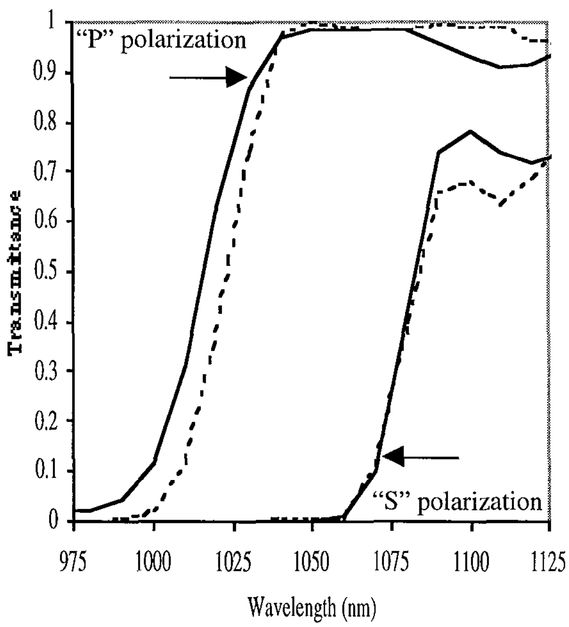


Fig.7 Spectral performance of Brewster's angle plate polarizers deposited from hafnia (dotted line) or hafnium (solid line) and silica.

5. CONCLUSIONS

The quality of hafnia silica multilayer thin films can be improved by evaporating Hafnium instead of hafnia in an electron-beam deposition process. This process modification results in a 3-10× defect reduction, improved interfaces, and 3× reduction in plume instability. The impact of these improvements is less conditioning-induced surface changes, elimination of flat-bottom pit laser damage, and improved spectral performance. This process improvement will be used to manufacture high laser fluence resistant large-aperture polarizer and mirror coatings for the National Ignition Facility.

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