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Effectiveness of ion cleaning to improve the laser damage threshold of HfO_2/SiO_2 optical coatings for high reflection and antireflection at 527 nm and 1054 nm

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ABSTRACT

Preventing contamination is vital to achieving high laser-induced damage thresholds in optical coatings. The importance of removing contamination from optical substrates has led to the development of many specialized cleaning processes, including the application of solvents, acids, mild detergents, and abrasives. To further enhance contamination removal, the substrate may be treated with ion cleaning just prior to depositing the optical coating. Ion cleaning is attractive thanks to the convenience of providing in-situ treatment to optical substrates, and also avoiding the hassle of managing hazardous chemicals or applying mechanical force to scrub off detergents and other cleaning agents. In this study, we compare the effectiveness of ion cleaning for increasing the laser-induced damage thresholds of high reflection (527 nm and 1054 nm) and antireflection (527 nm) coatings. Ion cleaning was performed using a radio frequency ion source with argon and oxygen. The coatings investigated were deposited with layers of HfO_2 and SiO_2 in an e-beam evaporation system, and are designed to withstand nanosecond pulses from a kJ-class laser.

Keywords: optical coatings, ion cleaning, HfO_2, SiO_2, laser damage, high reflection, antireflection

1. INTRODUCTION

The large optics coating system[1] at Sandia National Laboratories has been in operation since 2005, and uses e-beam evaporation of coating materials in a 2.3 X 2.3 X 1.8 m vacuum coating chamber to provide optical coatings with high resistance to laser damage for the Z-Backlighter laser system[2]. These lasers are kJ-class, pulsed systems operating with ns pulses in the terawatt range (527 nm), and ns to sub-ps pulses in the petawatt range (1054 nm). They provide an important backlighting diagnostic for Sandia's Z-machine, the most powerful and efficient laboratory radiation source in the world[3].

The cleanliness of an optical substrate is an important factor in realizing a high laser-induced damage threshold (LIDT), and we use a cleaning process that involves manually scrubbing the optical substrates with deionized water, mild detergent (Micro 90), and a mild abrasive (Baikalox)[4]. This manual cleaning process has been very effective, but we wanted to investigate additional cleaning methods because the LIDTs of some of our optical coatings have been negatively affected by the presence of defects, possibly on the optical substrates[5,6]. An in-situ cleaning processes such as ion cleaning with oxygen or argon is a convenient way to supplement our existing manual cleaning process and further reduce the presence of defects on optical substrates. Therefore, in this study we tested the effectiveness of an ion cleaning process for improving the LIDTs of some of our most common optical coatings.

2. METHOD

Ion cleaning of optical substrates was conducted with a 16-cm diameter, 3-grid, radio-frequency ion source (Veeco), shown in operation in Fig. 1. The ion source is directed at an angle of about 55 degrees and aims towards the center of the rotating planet cans that hold optical substrates. This is the same ion source that we use on a regular basis for ion-assisted deposition[1, 7-9], and it has also been used for ion etching to completely remove optical coatings from substrates[10].

Figure 1. The ion source in operation within the coating chamber.

The in-situ optical substrate ion cleaning method that we tested in this study has similar parameters compared to the previous ion etch process[10], however the ion etch parameters were aggressive and caused the formation of defects on the optical substrate. For this reason, we selected process parameters for ion cleaning that were more gentle compared to our selection for ion etching. The ion cleaning process parameters are shown below in Table 1, and it is a 2-step process involving ionized oxygen followed by ionized argon. The oxygen ions are favorable to cleaning through chemical reactions that break down organic contamination, while the argon ions are favorable to cleaning through mechanical means via the bombardment and removal of surface contamination. Using this type of 2-step ion cleaning process that involves two different cleaning mechanisms (chemical reactions and physical bombardment) is a promising strategy to ensure the removal of both organic and inorganic contamination.

Table 1. Ion Source Parameters for Ion Cleaning

Step	**Duration (min.)**	**Beam current (mA)**	**Beam voltage (V)**	**Ar gas flow (sccm)**	**O_2 gas flow (sccm)**	**Ar neutralizer flow (sccm)**
1	10	500	400	3	52	5
2	5	400	400	40	0	8

The ion source parameters of this study may not be applicable to other coating chambers due to the many different sizes and configurations of coating chambers and ion sources.

Our optical coatings with high resistance to laser damage consist of alternating HfO_2 and SiO_2 thin film layers. The HfO_2/SiO_2 optical coating designs that we selected to test in this study are listed below, and they represent some of our most common coatings. All of the optical substrates were 50-mm diameter, 10-mm thick, fused silica optically polished substrates that were cleaned using our standard, manual cleaning method[4]. All of the coatings were then produced with and without the in-situ ion cleaning process prior to deposition.

- Antireflection (AR), 527 nm, 0^0 angle of incidence (AOI)
- High reflection (HR), 527 nm, 45^0 angle of incidence (AOI), P-polarization
- High reflection (HR), 1054 nm, 45^0 angle of incidence (AOI), P-polarization
- (The AR coating consists of 4 layers; all HR coatings are 34-layer quarter-wave stacks with ½-wave outer SiO_2 layer)

For this study, the SiO_2 layers were produced from the evaporation of SiO_2 granules approximately 3 mm in size, with a deposition rate of 7 Å/s. The HfO_2 layers were produced from the evaporation of Hf metal in an oxygen environment (1.1e-4 Torr total pressure in the coating chamber), with a deposition rate of 3 Å /s. The deposition temperature was 200

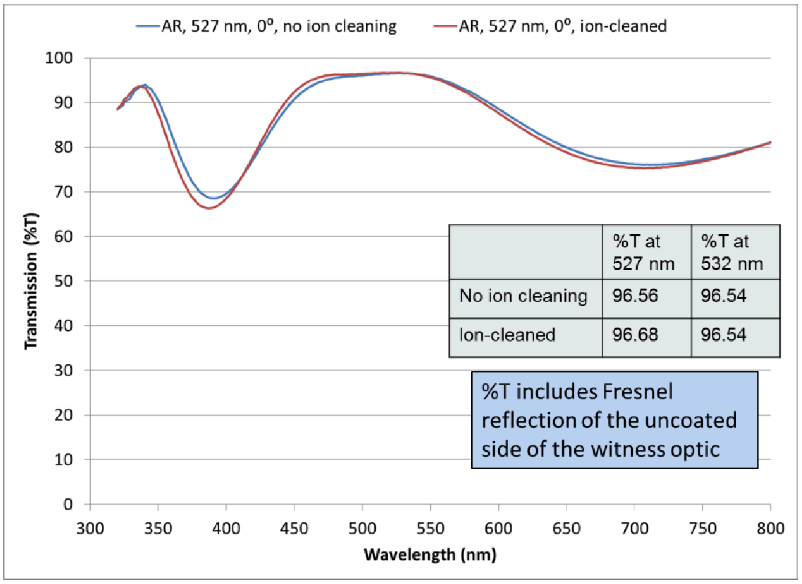
^{0}C. A single quartz crystal monitor is used to monitor the deposition rate, and masking provides a coating uniformity of +/- 0.5% on substrates as large as 94 cm in length[11]. There are 3 optical substrate holders called planet cans that rotate around a center gear during deposition.

The laser damage measurements were conducted by Spica Technologies, Inc.[12] using the NIF-MEL method[13]. In this protocol, the coated surface of the test optic first undergoes an alcohol drag-wipe cleaning step. Then, single transverse mode, multi-longitudinal mode laser pulses of 3.5 ns duration and produced at a 5 Hz repetition rate in a 1 mm diameter collimated beam are incident one at a time per site in a raster scan composed of ~ 2500 sites over a 1 cm^2 area. In the raster scan, the laser spot overlaps itself from one site to the next at 90% of its peak intensity radius. The laser fluence typically starts at 1.0 J/cm^2 in the cross section of the laser beam. After testing the 2500 sites at 1 J/cm^2, the fluence is increased in a 3.0 J/cm^2 increment and the 2500 sites are tested again. This progression repeats until the damage threshold fluence is reached.

The NIF-MEL procedure is essentially an N-on-1 test at each of the 2500 sites. Laser damage is identified as some type of melt or crater that alters the coated surface, but in some cases the damage stabilizes as a damage site that does not propagate (does not grow in size) as the laser fluence increases. In other cases, the damage does propagate. According to the NIF-MEL damage criterion, the LIDT is reached at the fluence at which 1 or more propagating damage sites occurs, or the fluence at which the number of non-propagating (NP) damage sites accumulates to at least 25, whichever fluence is the smaller. The 25 or more NP sites are 1% or more of the 2500 sites tested and constitute about 1% or more of the 1 cm^2 coating area tested. Our reasoning behind this LIDT criterion is the following. We know we cannot tolerate a propagating damage site in the laser beam train because it will quickly develop into catastrophic damage in the form of a large crater in the optic or worse; and 25 or more NP damage sites per cm^2, while they are benign because they do not grow, are flaws in the coating that scatter about 1% or more of the laser light out of the beam, and that level of loss of laser intensity is unacceptable for us.

3. RESULTS

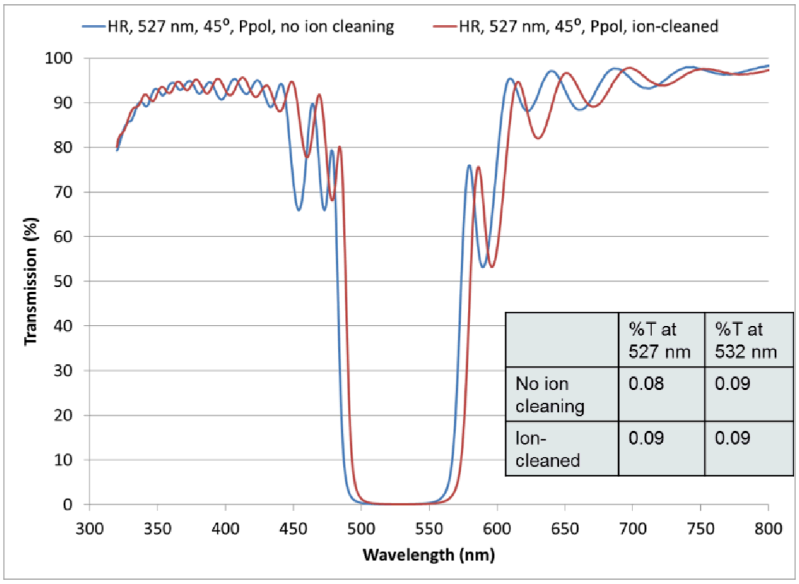
The transmission spectral scans of each coating, with and without ion-cleaning, are shown in Figs. 2-4. The spectral scans were acquired using a Perkin-Elmer Lambda 950 spectrophotometer. Transmission differences between the coatings are likely not affected by substrate ion cleaning, and instead reflect subtle process variations between the subsequent coating depositions. While the coatings are intended for operation at either 527 nm or 1054 nm, the LIDT tests were conducted at the closest available wavelengths (532 nm and 1064 nm). Figures 2-4 therefore show comparisons of the coatings' transmission at 527 nm and 532 nm, or 1054 nm and 1064 nm. Electric field differences in the coatings are assumed to be negligible between 527 nm and 532 nm, and 1054 nm and 1064 nm.



	%T at 527 nm	%T at 532 nm
No ion cleaning	96.56	96.54
Ion-cleaned	96.68	96.54

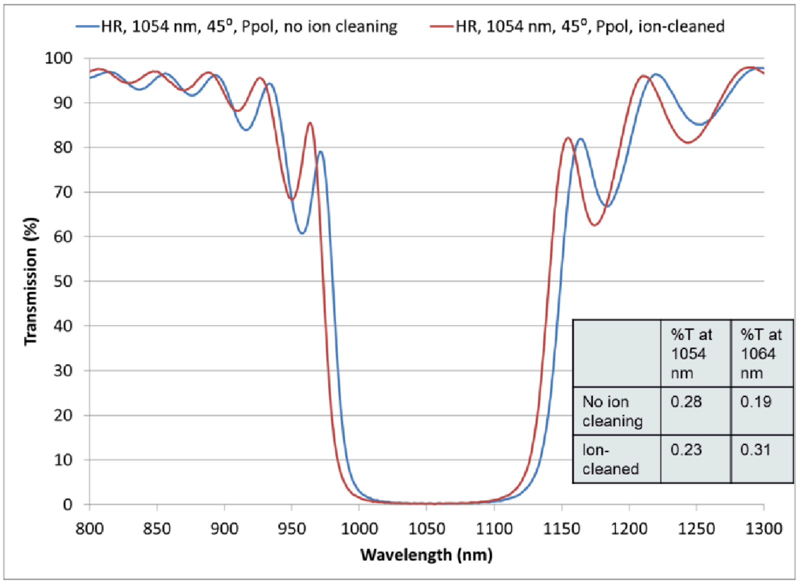
%T includes Fresnel reflection of the uncoated side of the witness optic

Figure 2. Transmission spectral scans of the AR coatings for 527 nm.



	%T at 527 nm	%T at 532 nm
No ion cleaning	0.08	0.09
Ion-cleaned	0.09	0.09

Figure 3. Transmission spectral scans of the HR coatings for 527 nm.



	%T at 1054 nm	%T at 1064 nm
No ion cleaning	0.28	0.19
Ion-cleaned	0.23	0.31

Figure 2. Transmission spectral scans of the HR coatings for 1054 nm.

The LIDTs of each coating, with and without substrate ion cleaning, are shown in Fig. 5. Unfortunately, the ion cleaning process that we selected did not help to improve the laser damage thresholds of any of the coatings and, in the case of two coatings (AR for 527 nm, and HR for 1054 nm), the LIDTs in the ion-cleaned cases actually decreased.

In the HR coatings, very little laser light actually reaches the substrates. However, the AR coatings are very transmissive and most of the laser light reaches the substrates, which could interact with substrate contamination/defects and cause damage. The fact that neither the AR nor HR coatings saw LIDT improvements with substrate ion cleaning indicates that the condition of the substrate may not have influenced the LIDT, or perhaps ion cleaning was not effective and the condition of all the substrates remained about the same, whether they were ion cleaned or not. In later research, we can examine the damage morphology of these samples to confirm our claims, and attempt to optimize the ion cleaning process.

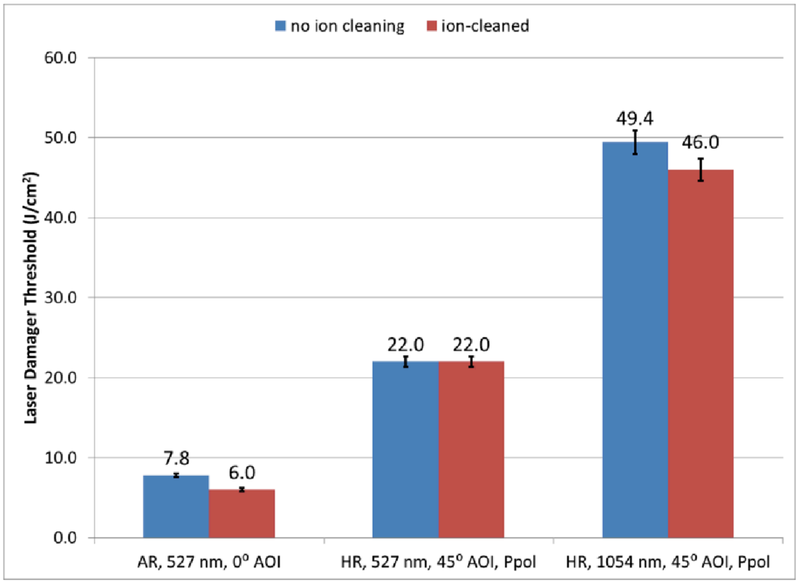


Figure 5. LIDTs of all coatings, with and without substrate ion cleaning.

The number of non-propagating (NP) damage sites detected in each coating during LIDT testing is plotted in Fig. 6. There is not a clear relationship between substrate ion cleaning and whether the LIDTs were established due to propagating or non-propagating damage. Two of the coatings (AR for 527 nm, and HR for 1054 nm) behave similarly, in that the number of NP damage sites detected in both the ion-cleaned and non-ion-cleaned samples follows trends that almost overlap. On the other hand, the NP damage sites detected in the HR coatings for 527 nm do not follow a similar trend. In fact, much fewer NP damage sites were detected in the ion-cleaned sample. However, the reduction of NP damage in the ion-cleaned sample probably cannot be attributed to ion cleaning because this trend was not observed in the data of the other two coatings. It is more likely instead that slight process variations between each coating deposition are responsible for the LIDT differences between ion-cleaned and non-ion-cleaned samples. We have seen this before in HR coatings for 527 nm[5]. Another way to view this is that any benefits of the ion cleaning process for improving LIDTs could have been overshadowed by the more dominating influence of defects caused by coating deposition variations and resulting in NP damage. However, as stated before, it is also possible that the ion cleaning process was ineffective, and this is why the LIDTs did not improve, and were instead influenced by coating process variations or contamination that still remained on the substrate.

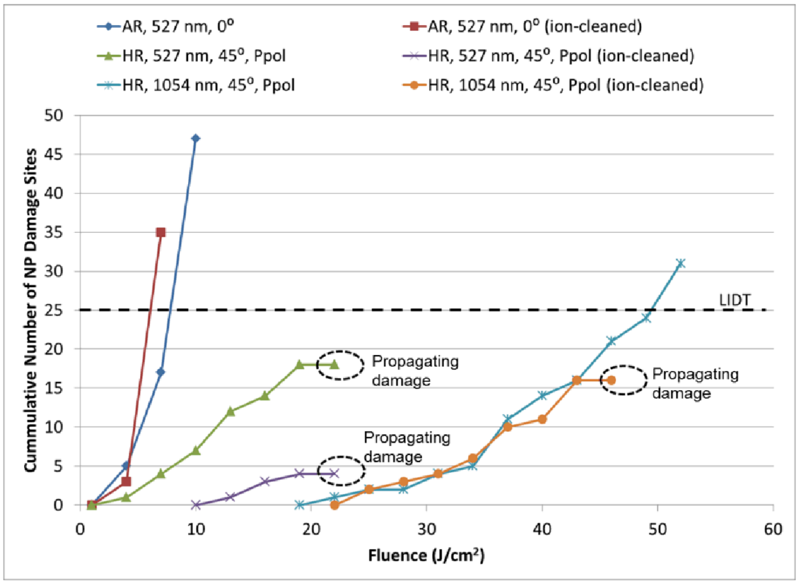


Figure 6. The number of NP damage sites detected in each coating during LIDT testing. The black, dashed outlines show samples that reached their LIDT due to propagating damage. All other samples reached their LIDT due to the presence of >25 NP damage sites.

4. CONCLUSION

In this study, we tested an in-situ, ion cleaning process for optical substrates to determine whether this could further improve the LIDTs of our HfO_2/SiO_2 optical coatings for 527 nm and 1054 nm. Unfortunately, the coated samples that received substrate ion cleaning did not exhibit improved LIDTs. This indicates that substrate ion cleaning may not be a necessary treatment for our optics, but further investigation is needed. It is also possible that the ion cleaning method we selected was not effective, and therefore the condition of all the substrates remained about the same, whether they were ion-cleaned or not. It is worth experimenting with different ion source parameters to determine the most effective settings for the types of contamination that we deal with.

The LIDTs of most coated samples were dominated by non-propagating damage. The source of defects responsible for this non-propagating damage will need to be investigated more directly in order to improve LIDTs.

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