SAND2018-12722C

The impact of contamination and aging effects on the long term laser damage resistance of SiO_2/HfO_2/TiO_2 high reflection coatings for 1054 nm

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ABSTRACT

The laser damage thresholds of optical coatings can degrade over time due to a variety of factors, including contamination and aging. Optical coatings deposited using electron beam evaporation are particularly susceptible to degradation due to their porous structure. In a previous study, the laser damage thresholds of optical coatings were reduced by roughly a factor of two from 2013 to 2017. The coatings in question were high reflectors for 1054 nm that contained SiO_2 and HfO_2 and/or TiO_2 layers, and they were stored in sealed PETG containers in a class 100 cleanroom with temperature control. At the time, it was not certain whether contamination or thin film aging effects were responsible for the reduced laser damage thresholds. Therefore, to better understand the role of contamination, the coatings were recleaned and the laser damage thresholds were measured again in 2018. The results indicate that contamination played the most dominant role in reducing the laser damage thresholds of these optical coatings, even though they were stored in an environment that was presumed to be clean.

Keywords: Optical Coatings, Hafnia, Silica, Titania, Laser Damage, Mirrors, Aging, Contamination

1. INTRODUCTION

Optical coatings with high resistance to laser damage are integral to the operation of the Z-Backlighter laser facility [1] at Sandia National Laboratories. The Z-Beamlet kJ-class laser operates with ns pulses in the terawatt peak power range at both 1054 nm and 527 nm, while the Z-Petawatt laser operates with ns to sub-ps pulses at 1054 nm. Our large optics coating system [2] utilizes electron beam evaporation to produce optical coatings that usually consist of hafnia (HfO_2) and silica (SiO_2) layers to achieve high laser-induced damage thresholds (LIDTs).

In previous studies [3,4,] we experimented with replacing hafnia layers with titania (TiO_2) layers in mirror coatings to increase high reflection bandwidth and angle-of-incidence (AOI) flexibility at 1054 nm. However, titania has a lower bandgap and exhibits lower LIDTs compared to hafnia [5]. We also discovered that the LIDTs of both hafnia and titania-based mirror coatings from 2013 degraded even more after the coatings were stored for 4 years in sealed PETG containers in a temperature-controlled, class 100 clean room environment [6]. At the time, it was unclear whether contamination and/or aging were responsible for the reduced LIDTs, although both phenomena were suspected [6]. Therefore, the aim of this study was to better understand how contamination and aging influenced the lower LIDTs. In a nutshell, this was accomplished by cleaning the coatings, and then measuring the LIDTs again to determine if laser damage resistance was improved by the cleaning.

2. METHOD

The mirror coatings that were tested in this study were first produced in 2013 [3]. They are all 42-layers, quarter-wave stack designs, for 1054 nm, 45° AOI, P-polarization, with a half–wave outer layer of silica to improve resistance to laser damage [7]. Silica was used for the low index layers, and hafnia and/or titania were used for the high index layers. The model in Fig. 1 shows that electric field intensities diminish near the substrate. The titania layers replaced hafnia layers near the substrate to afford more protection from the high electric field intensities, since titania has a lower LIDT compared to hafnia.

Of the five coatings produced in 2013, they contained either 7, 10, 13, 16, or 21 inner titania layers, with the coating containing 21 titania layers having no hafnia layers. An example of the layer thicknesses of the coating containing

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10 inner titania layers is shown in Fig. 2. Also, a sixth coating was added to this study at a later time: in 2014, we produced a coating of the same design containing just hafnia and silica layers to use for comparison.

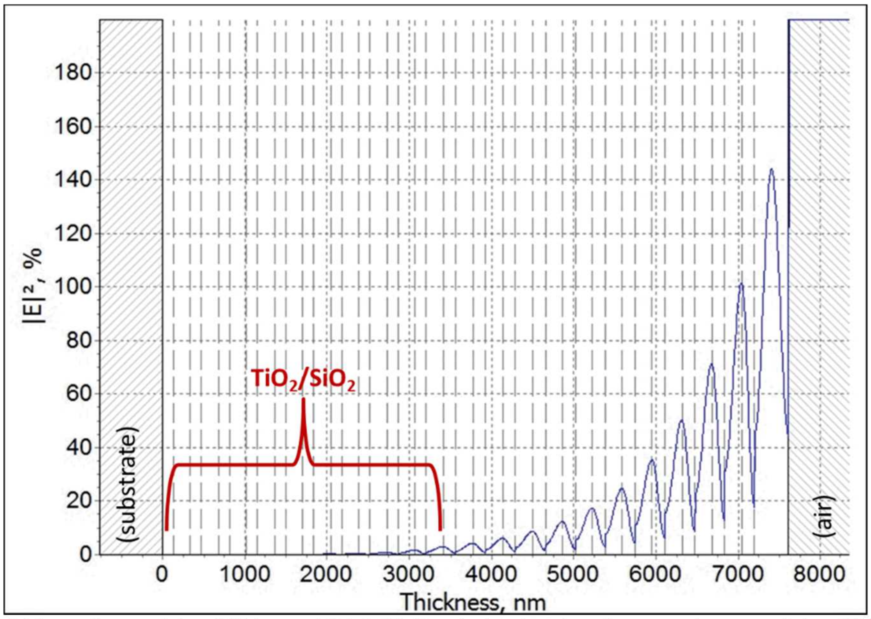

Figure 1: Electric field intensity model at 1054 nm, 45□ AOI, P-polarization, in mirror coating containing 10 inner titania layers and 11 outer hafnia layers. Vertical dashed lines indicate layer boundaries. The electric field intensity diminishes near the substrate. Using the titania layers near the substrate in the area of lowest electric field intensity is a strategy to protect titania layers from laser damage. This model was generated with Optilayer software [8].

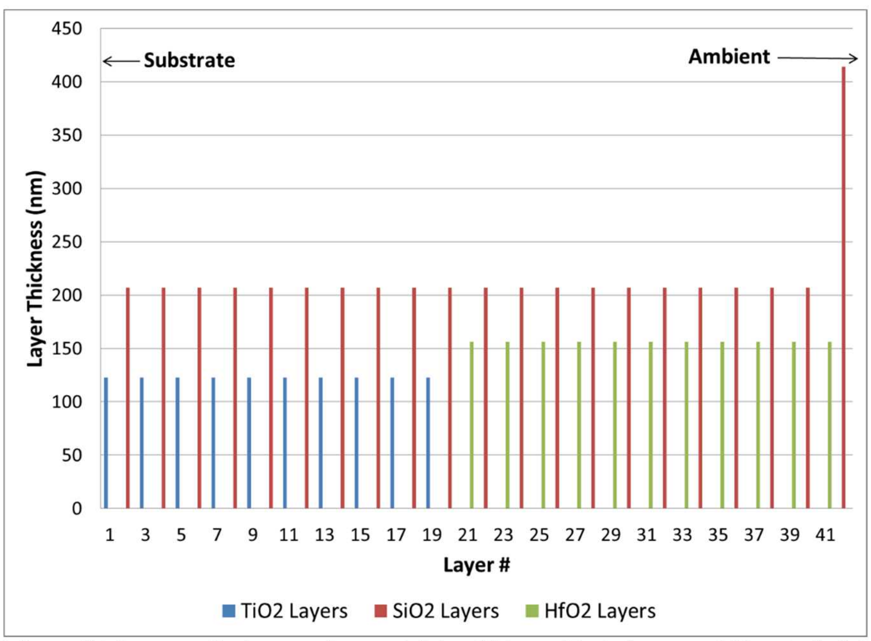

Figure 2: Layer thicknesses of mirror coating containing 10 inner titania layers and 11 outer hafnia layers.

Each coating from 2013 was deposited on an optically polished fused silica substrate, 2” diameter X 0.5” thick. The hafnia/silica coating produced in 2014 was deposited on a BK7 substrate. Prior to coating, each substrate was cleaned using our standard process [9], which involves Micro 90 detergent, Baikalox alumina slurry, and deionized water.

The coating process was electron beam evaporation in our custom chamber [2], with ion-assisted deposition (IAD) from a 16-cm diameter RF ion source. The coating parameters are shown below in Table 1. In addition to IAD, the hafnia and titania layers were deposited with a backflow of oxygen gas into the coating chamber. The total pressure in the coating chamber during the deposition of hafnia and titania layers in 2013 was 1.5e-4 Torr, but this pressure measurement is suspect because our ion gauge was not calibrated at the time. In 2014, using a calibrated ion gauge, the total pressure in the coating chamber was 1.2e-4 Torr during deposition of the hafnia layers. The deposition temperature was 200° C for all coatings. In addition, the coating system used masking to maintain uniformity, planetary rotation, and quartz crystal monitoring for layer thickness control.

Table 1: Coating Deposition Parameters with IAD

	Starting material	**Deposition rate (Å/s)**	**Ion beam current (mA)**	**Ion beam voltage (V)**	**Ar neutralizer flow (sccm)**	**Ar flow (sccm)**	**O_2 flow (sccm)**
SiO_2	1-3 mm granules	7	425	400	6	25	25
HfO_2	Hf shavings	3	600	400	7	0	45
TiO_2	Ti 2-3 mm granules	3	600	400	7	0	45

Following deposition, each coated sample was cleaned. Previous studies have shown that LIDTs can degrade due to environmental factors such as contamination, but in some cases the LIDT recovered to the original values after the coatings were cleaned [10, 11] or baked [12]. However, our lab is not equipped to accommodate the cleaning and bakout processes described in [10-12] if they are applied to meter-class optics. Therefore, we utilized our standard cleaning process instead, because it scales up to cleaning meter-class optics with our existing equipment. Our standard cleaning process involves washing the optic by hand with deionized water and Micro 90 detergent [9].

Following cleaning, all samples were sent to Spica Technologies [13] for initial LIDT testing in 2013 or 2014. Since then, all samples were stored in clean polyethylene terephthalate –G (PETG) plastic containers in our class 100 clean room, which is temperature controlled. When LIDT testing occurred again in 2017, the samples were not cleaned, and testing was performed in a different quadrant of the sample compared to before. The results in 2017 revealed that the LIDTs of each coating diminished significantly, often by a factor of 2, and this LIDT decline occurred regardless of the number of titania layers in the coating [6]. Because contamination was a suspected cause of the LIDT decline, in 2018 the samples were cleaned with Micro 90 detergent and deionized water, and sent to Spica Technologies again for laser damage testing in a different quadrant. In total, each sample has undergone 3 LIDT tests: 2013 or 2014 (after cleaning), 2017 (no cleaning), and 2018 (after cleaning).

The LIDTs were measured at 1064 nm, 45° AOI, in P-polarization. The LIDT tests were conducted according to the NIF-MEL protocol [14]. Single transverse mode (Gaussian), 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 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 1 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 – that is, grow in size – as the laser fluence increases. These non-propagating (NP) damage sites tend to be caused by the interaction of the laser field with nano-defects (pits, nodules, or contamination) in the coating. In other cases, the damage does propagate. Propagating damage tends to be intrinsic, governed by how the laser field interacts directly with the coating molecules.

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 NP damage sites accumulates to at least 25, whichever fluence is 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 reason for choosing an LIDT test with these damage criteria 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 may not grow, are flaws in the coating that scatter about 1% of the laser light out of the beam, and that level of loss of laser intensity is unacceptable for us.

The initial LIDT tests conducted in 2013/2014 were performed in the ambient environment (that is, some humidity was present), while the tests conducted in 2017 and 2018 were performed in a dry nitrogen environment (0% humidity present).

3. RESULTS

The spectral transmission characteristics of each coating are shown in Fig. 3. The coating containing 21 titania layers (0 hafnia layers) has an HR bandwidth of 232 nm, while the coating containing 0 titania layers (21 hafnia layers) has an HR bandwidth of 77 nm (HR bandwidth is taken as interval where transmission < 0.5%). Between 2013 and subsequent measurements in 2017, the coatings experienced a spectral shift due to aging, and the 2017 measurement of the coating containing 21 titania layers is shown in Fig. 3 as an example. The high reflection band of this coating was originally centered at 1088 nm and is now centered at 1119 nm. There is a spectral shift of 31 nm, which is 2.85% larger than the original 1088 nm bandwidth. However, the HR bandwidth of this coating still encompasses the LIDT test wavelength of 1064 nm.

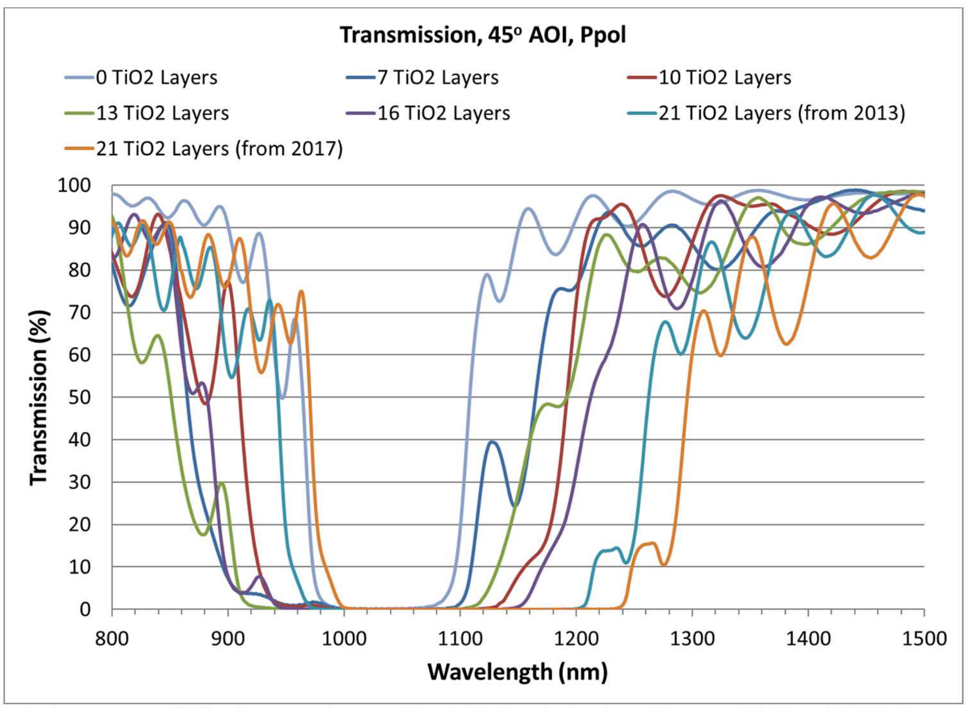

Figure 3: Transmission spectra of all mirror coatings at 45□ AOI, P-polarization. Between 2013 and 2017, the coatings experienced a spectral shift, and the coating containing 21 titania layers is shown as an example.

The spectral shift to longer wavelength is not a surprise. Due to their porosity, coatings deposited with electron beam evaporation undergo a spectral shift to longer wavelength, often referred to as 'aging.' Aging mostly occurs within the first month of deposition. This means the center of the HR bandwidths in 2013 are slightly lower than those in 2017, and this hampers our ability to make a straightforward comparison between the initial LIDT tests and the LIDT tests that occurred in 2017 and 2018. Furthermore, the difference in humidity between the initial LIDT tests (ambient humidity) and the 2017/2018 tests (0% humidity) also limit our ability to directly compare LIDT results. Therefore, the LIDT results from 2013 are suitable mostly as an approximation. In 2013, if we had the foresight to measure the LIDTs in a dry environment, the initial LIDT values would be a more reliable reference.

The LIDT of each coating is presented in Fig. 4. These results indicate that LIDTs can be improved significantly by cleaning the coatings with mild detergent after they have spent years in storage. This suggests contamination can accumulate in storage after long periods even if the storage method was presumed to be clean (sealed PETG containers, class 100 clean room, temperature control). The fact that the LIDTs improved after cleaning with mild detergent and deionized water indicates that most of the contamination was merely on the surface. It is also interesting that something as simple as surface contamination can be so detrimental to optical coatings: in this case the LIDTs were reduced by a factor of 2 or more regardless of the number of titania layers.

The LIDTs from 2018 did not recover to their original values. However, as mentioned earlier, the original LIDT values should be understood as approximations, so we do not expect the LIDT measurements from 2018 to be an exact match. That being said, it is unclear how much the mismatch between the 2018 LIDT values and the original LIDT values is due to contamination that may have remained after the cleaning process.

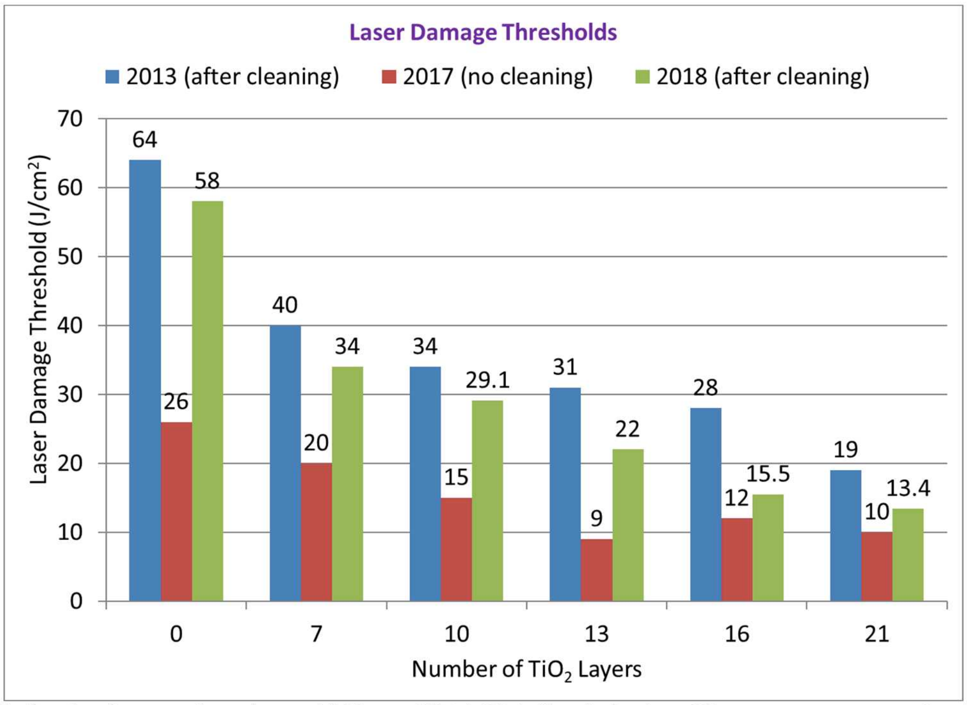

Figure 4: LIDT of each mirror coating taken at 1064 nm, 45□ AOI, in P-polarization. The measurement error is +/- 1 J/cm^2. The LIDT degradation between the initial tests and the later tests taken in 2017 is an apparent characteristic of every coating.

As mentioned previously, the NIF-MEL protocol tracks the number of non-propagating (NP) defects present in the coating at the damage fluence. The number of NP defects observed for each coating are shown in Fig. 5. The initial LIDT tests have an NP defect distribution that appears random, regardless of the number of titania layers in the coating. However, the additional LIDT data from 2017 and 2018 indicate a trend towards higher numbers of defects in coatings with more titania layers. Interestingly, cleaning the coatings in 2018 in most cases did not reduce the number of defects present, even though laser damage thresholds improved. This suggests that contamination is not fully responsible for NP defects. Intrinsic properties that change over time, particularly in the coatings that contain more titania layers, may be responsible for the escalation of NP defects.

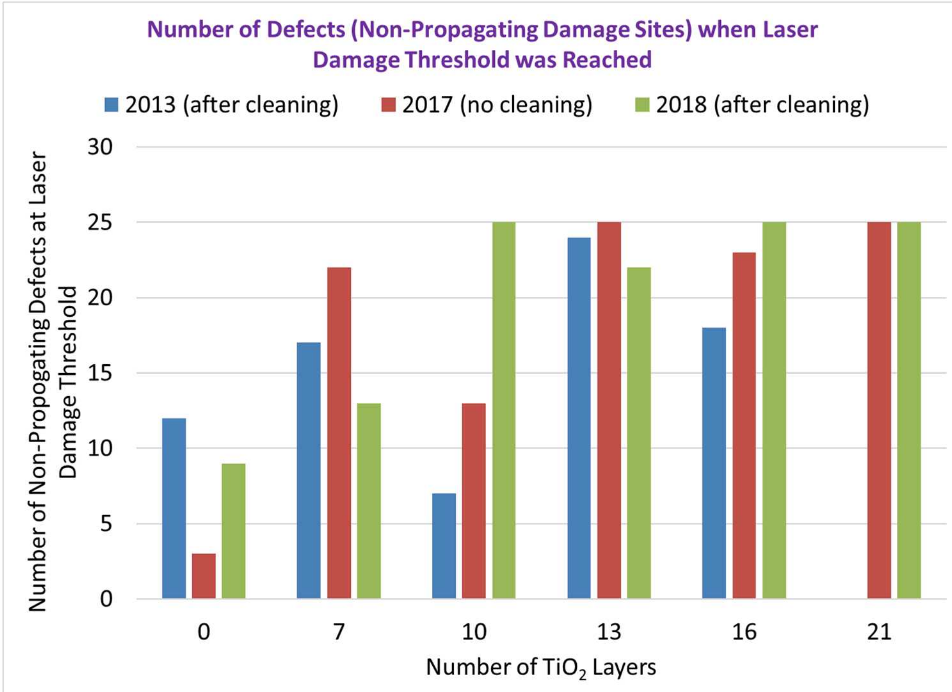

Figure 5: Number of NP defects present at the laser damage threshold fluence in each mirror coating.

4. CONCLUSION

Hafnia/titania/silica mirror coatings that were inadvertently exposed to surface contaminants during storage exhibited lower LIDTs. These diminished LIDTs were improved by cleaning the coatings with mild detergent and deionized water. However, while improved LIDTs were observed, coatings containing higher numbers of titania layers exhibited the highest numbers of non-propagating defects, and in most cases cleaning did not remedy this. It was therefore considered that the defects in the titania films may have emerged due to intrinsic properties that changed over time. Further studies could be devoted to improving the quality of the titania films to decrease the growth of defects. Also, if the LIDT tests had been conducted in the femtosecond regime, this could have helped to identify the impact of intrinsic thin film properties on the reduction of LIDTs over time, since ultrafast LIDTs depend mainly on intrinsic properties (bandgap) of the coating materials rather than extrinsic problems such as contamination [15, 16].

ACKNOWLEDGEMENTS

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia LLC, a wholly owned subsidiary of Honeywell International Inc. for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525. This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

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