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Laser Damage Comparisons of E-Beam Evaporated HfO_2/SiO_2 Antireflection Coatings at 0% and 40% Relative Humidity for 532 nm and 1064 nm

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ABSTRACT

Antireflection coatings, containing alternating layers of hafnia (HfO_2) and silica (SiO_2), were deposited using electron beam (e-beam) evaporation for use in laser operations at 532 nm and 1064 nm in the nanosecond regime. The e-beam evaporation process produces coatings that are porous and therefore absorb water from the ambient environment. Consequently, humidity may affect the spectral performance of the coatings, and the laser damage resistance of the coatings may be affected as well. The purpose of this study was to compare the laser-induced damage thresholds of the antireflection coatings measured in the ambient environment at 0% and 40.5% relative humidity. At 1064 nm, the laser-induced damage thresholds at 0% and 40.5% relative humidity were almost the same. However, at 532 nm, the laser-induced damage thresholds at 40.5% relative humidity were nearly twice as high as those measured at 0% relative humidity. This indicates that humidity can inhibit lower-fluence precursors that would lead to laser damage at 532 nm in the nanosecond regime, thereby improving the durability of the coatings in a humid environment.

Keywords: Laser Damage, Optical Coatings, Humidity, Hafnia, Silica, E-Beam Evaporation

1. INTRODUCTION

The Z-Backlighter Laser Facility at Sandia National Laboratories is a kJ-class system utilizing optics up to a meter in size [1, 2]. Most of the optics have been coated with our own e-beam evaporation system and have high resistance to laser damage in the nanosecond regime at 532 nm and 1064 nm [2]. The coatings typically consist of alternating layers of HfO_2 and SiO_2.

For our facility, laser damage tests were typically conducted in a dry, 0% relative humidity environment, for the purpose of benchmarking the coatings under the same conditions. However, the coated optics are used in a variety of environments, such as vacuum, dry air, and humid ambient air. This is of interest because e-beam evaporation produces coatings that are porous and consequently absorb water from the ambient environment [3]. The absorbed water can impact the stress and spectral performance of a coating [4, 5]. Specifically, the absorbed water can cause a red shift which may affect the electric field distribution in the coating layers. There is also evidence from previous studies that humidity affects the laser-induced damage threshold (LIDT) of coatings produced by e-beam evaporation [3, 4, 6]. With this in mind, it was important to have our own coatings tested in different environments to understand the impact of humidity on the LIDT.

In this preliminary study, we chose to investigate antireflection (AR) coatings because their wide usage and lower LIDTs are an operational concern of our facility. In addition, it was assumed that the relatively simple design of the AR coatings would make it easier to distinguish the impact of humidity on the LIDT results. Ultimately, what we learned is that humidity did not improve nanosecond LIDTs at 1064 nm, but it delayed the onset of damage at 532 nm for these AR coatings.

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2. METHOD

In this study, two AR coatings for 532 nm and 1064 nm underwent laser damage testing at 0% and 40.5% relative humidity (RH). In this section, details about the coating design, deposition, and laser damage tests are presented.

The antireflection coatings consisted of 4 alternating layers of HfO_2 and SiO_2, and the layer thicknesses are shown in Fig. 1. The electric field distributions in the coating layers are shown in Figs. 2 and 3 for 532 nm and 1064 nm, respectively. The coating design and electric field models were generated using Optilayer software [7]. Based on the electric field models, at both 532 nm and 1064 nm, peak intensities occur in the SiO_2 layers. This is advantageous because SiO_2 has a higher LIDT compared to HfO_2. However, defects in the SiO_2 layers will also experience peak intensities. Furthermore, the conditions at 532 nm are a little worse since 532 nm is at a higher photon energy, and both SiO_2 layers experience peak intensities rather than just the outer SiO_2 layer. Therefore, we expect LIDTs to be lower at 532 nm compared to 1064 nm.

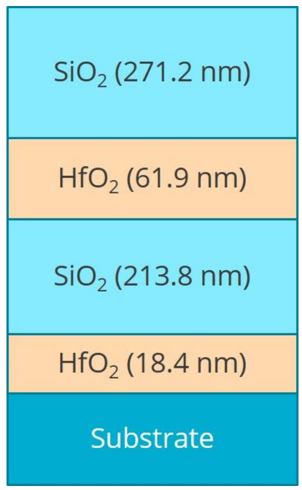

Figure 1. Layer thicknesses of antireflection coating consisting of alternating HfO_2 and SiO_2 layers.

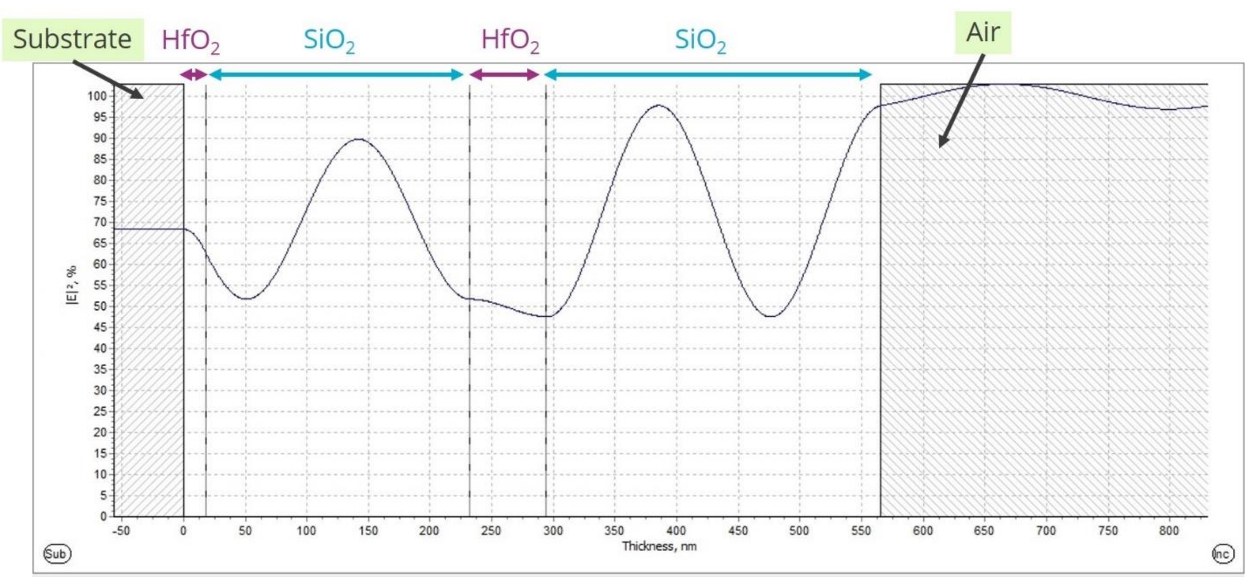

Figure 2. Model of electric field distribution at 532 nm and normal incidence. The highest electric field intensities are in both SiO_2 layers. The refractive indices of the coating materials at 532 nm were modeled as follows: $Re(n)_{HfO2} = 1.93$ and $Re(n)_{SiO2} = 1.45$.

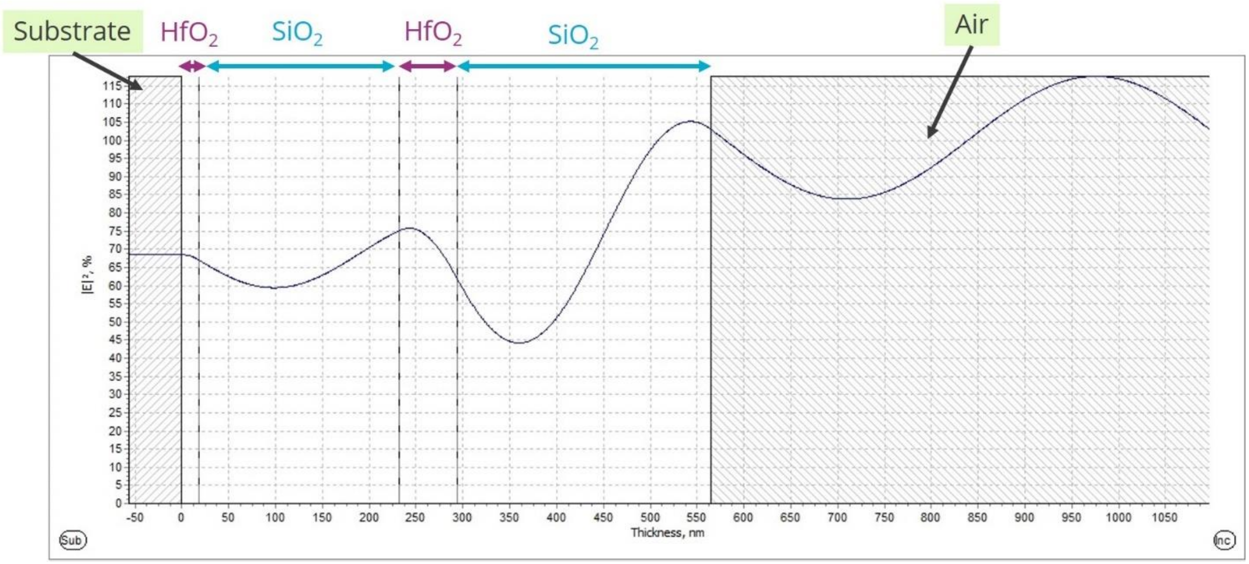

Figure 3. Model of electric field distribution at 1064 nm and normal incidence. The highest electric field intensity is in the outer SiO_2 layer. The refractive indices of the coating materials at 1064 nm were modeled as follows: $Re(n)_{HfO2} = 1.91$ and $Re(n)_{SiO2} = 1.44$.

The optical coating facility at Sandia National Laboratories is located in a class 100 clean room. The coatings were deposited on optically polished fused silica substrates (50 mm diameter, 12.7 mm thick). Just before the substrates were loaded into the coating chamber, they were cleaned using our standard process, which involves manually washing with mild Micro 90 detergent, Baikalox alumina slurry, deionized water, and clean room wipes (Texwipe TX1010) [8].

The coatings were deposited at a temperature of 200° C. The SiO_2 layers were deposited at a rate of 7 Å/s from 1-3 mm SiO_2 granules. The HfO_2 layers were deposited from hafnium metal with O_2 backfill, resulting in $1.3x10^{-4}$ Torr total pressure in the coating chamber [9]. The first HfO_2 layer was deposited at a rate of 1 Å/s, and the second HfO_2 layer was deposited at 2 Å /s. The coating system contains planetary rotation and masking to maintain uniformity, and layer thickness control was implemented with quartz crystal monitoring. After the coated substrates were removed from the coating system, they were allowed to age for almost 2 months before laser damage testing was conducted. Prior to laser damage testing, the substrates were cleaned with deionized water and Micro 90 detergent [8].

Specifically, two substrates were coated, with each one in a separate deposition using the design shown in Fig. 1. The flow chart shown in Fig. 4 illustrates the laser damage tests that were conducted on each coated substrate. In total, each coated substrate underwent 4 laser damage tests.

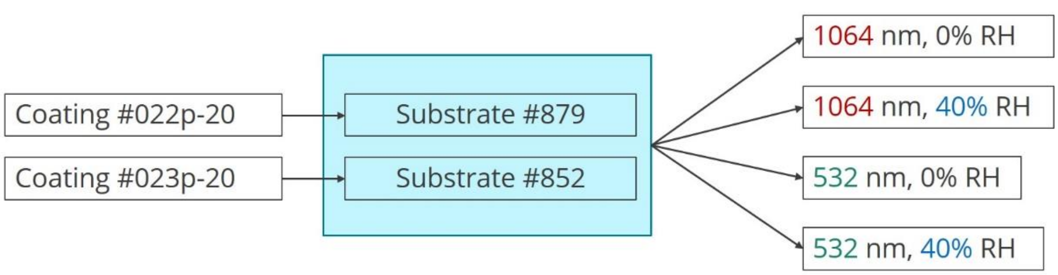

Figure 4. Flow chart of the 4 laser damage tests conducted on each coated substrate.

Laser damage testing was conducted by Spica Technologies using the NIF-MEL protocol [10, 11]. The tests were conducted at 0% and 40.5% relative humidity and normal incidence. Spica Technologies allowed each sample to reach a steady state with the environment by keeping the sample at the humidity condition for 12 hours before performing the laser damage test. The 0% RH condition was produced with dry nitrogen, and the 40.5% RH was produced with humidified air.

The NIF-MEL laser damage testing protocol is a raster-scan type test involving 2500 overlapping sites in a 1 cm^2 area. The test utilized 3.5 ns pulses with TEM_{00} beam profile and 5 Hz repetition rate. The spot sizes ($1/e^2$) were 1.01 mm (at 532 nm) and 1.04 mm (at 1064 nm). In the raster scan, consecutive laser spots overlap from site to site at the radius for 90% of the peak intensity. The laser fluence begins at 1 J/cm^2 at each of the 2500 sites, and is then increased by 1 J/cm^2 increments, repeating until the damage threshold fluence is reached. The accuracy of the fluence measurement is +/- 3% [10].

In the NIF-MEL protocol, laser damage is the fluence at which a propagating damage site occurs, or the fluence at which 25 or more non-propagating damage sites occur. The lowest fluence that satisfies either of those conditions is the LIDT. [11]

Propagating damage is generally associated with intrinsic properties of the coating, such as bandgap, which can lead to damage sites that grow catastrophically upon repeated irradiance [12]. Non-propagating damage is generally associated with coating defects such as nodules that were ejected from the coating upon irradiation of a certain fluence, resulting in a stabilized damage spot that does not grow upon subsequent irradiances [12]. In the nanosecond regime, laser damage is expected to be dominated by defects [13, 14].

3. RESULTS

The transmissions of both antireflection coatings are shown in Fig. 5 and were measured using a PerkinElmer Lambda 950 spectrophotometer. The transmission scans show that the coatings experienced a red shift of about 4 nm at 40% RH compared to 0% RH. However, this red shift of 4 nm is small and does not significantly offset the electric field distribution in the coating. To confirm, Optilayer was used to model the refractive indices of the coating #022p-20 at 0% and 40% RH based on the actual spectral transmission of the coating. As shown in Fig. 6, at 40% RH, the refractive indices of both HfO_2 and SiO_2 are slightly higher than those for 0% RH, which could be attributed to the higher index of the water content in the coating layers or changes in coating stress due to humidity. The refractive index change was applied to the electric field models, and Figs. 7 and 8 show that the slight increase in the refractive index at 40% RH has a negligible impact on the electric field distribution in the coating layers. Overall, these models suggest that any differences in LIDT between 0% and 40% RH probably cannot be attributed to the 4 nm spectral shift or changes in the electric field distribution.

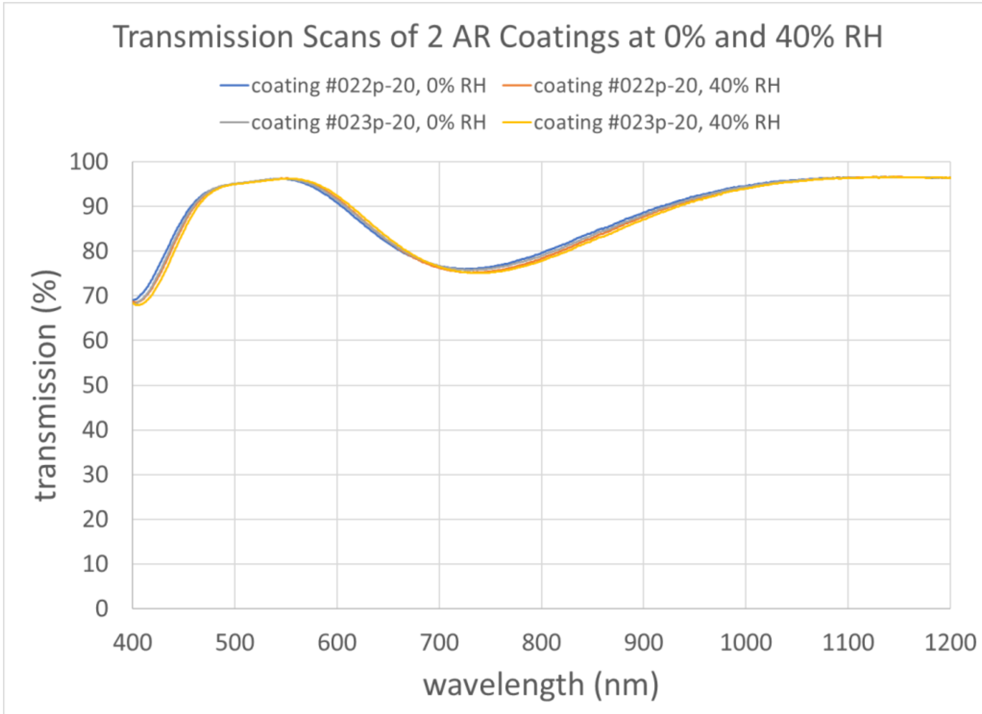

Figure 5. Transmission measurements at normal incidence of the antireflection coatings at 0% RH and 40% RH reveal that humidity caused a spectral shift of about 4 nm. (Note, the substrates were coated on one side, and therefore the transmission measurements include the Fresnel reflection from the uncoated side of the substrate).

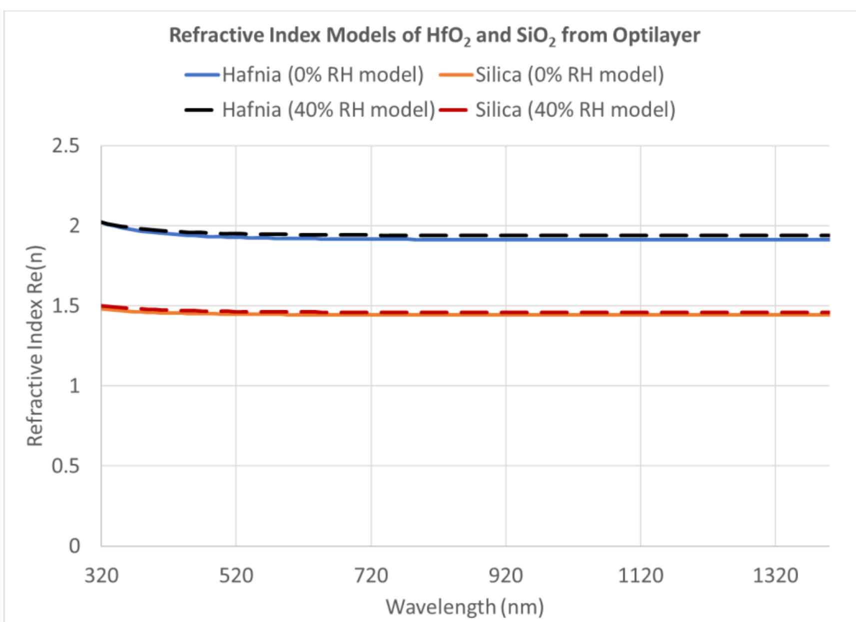

Figure 6. Refractive index models of the HfO_2 and SiO_2 coating materials at 0% and 40% RH. The models are based on the actual transmission data shown in Fig. 5. The modeled refractive indices at 40% RH are slightly higher than those at 0% RH.

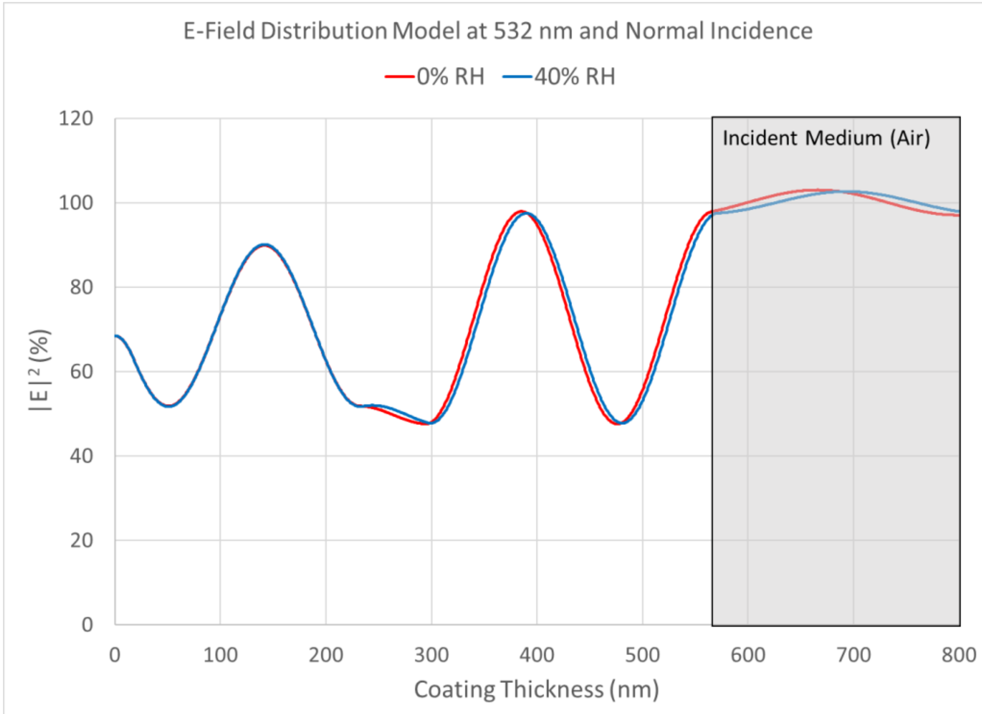

Figure 7. Electric field distribution models at 532 nm and normal incidence, showing minimal differences between 0% and 40% RH.

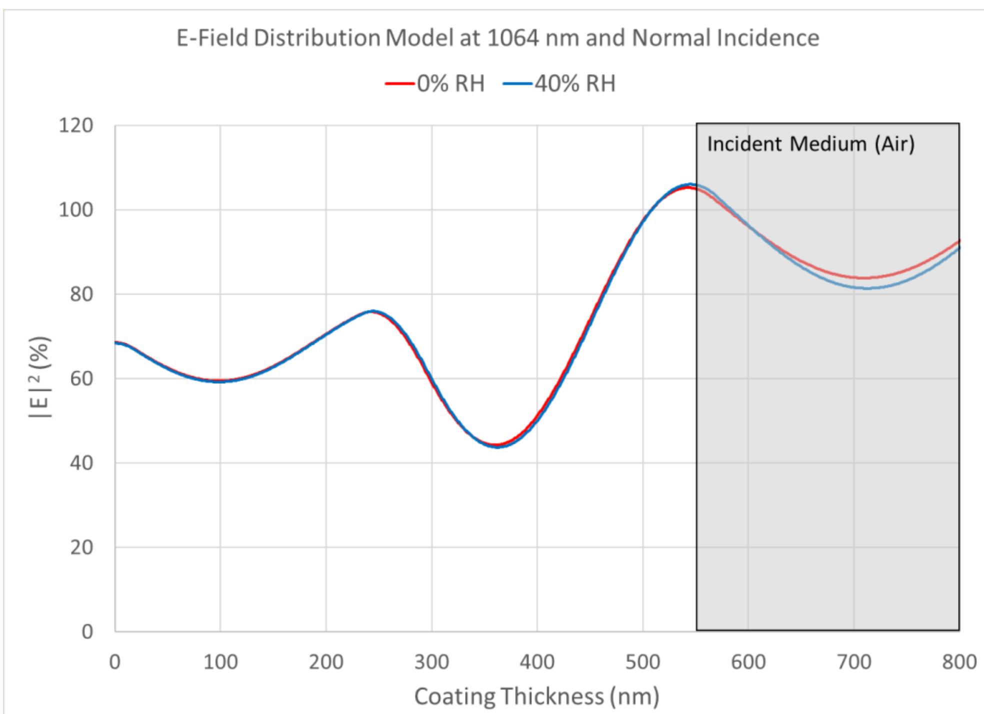

Figure 8. Electric field distribution models at 1064 nm and normal incidence, showing minimal differences between 0% and 40% RH.

The LIDT results are presented in Fig. 9. Based on the NIF-MEL damage criterion, at 532 nm, the LIDT increased by approximately twofold or threefold at 40.5% RH compared to 0% RH. However, at 1064 nm, the LIDT was nearly the same at 0% and 40.5% RH.

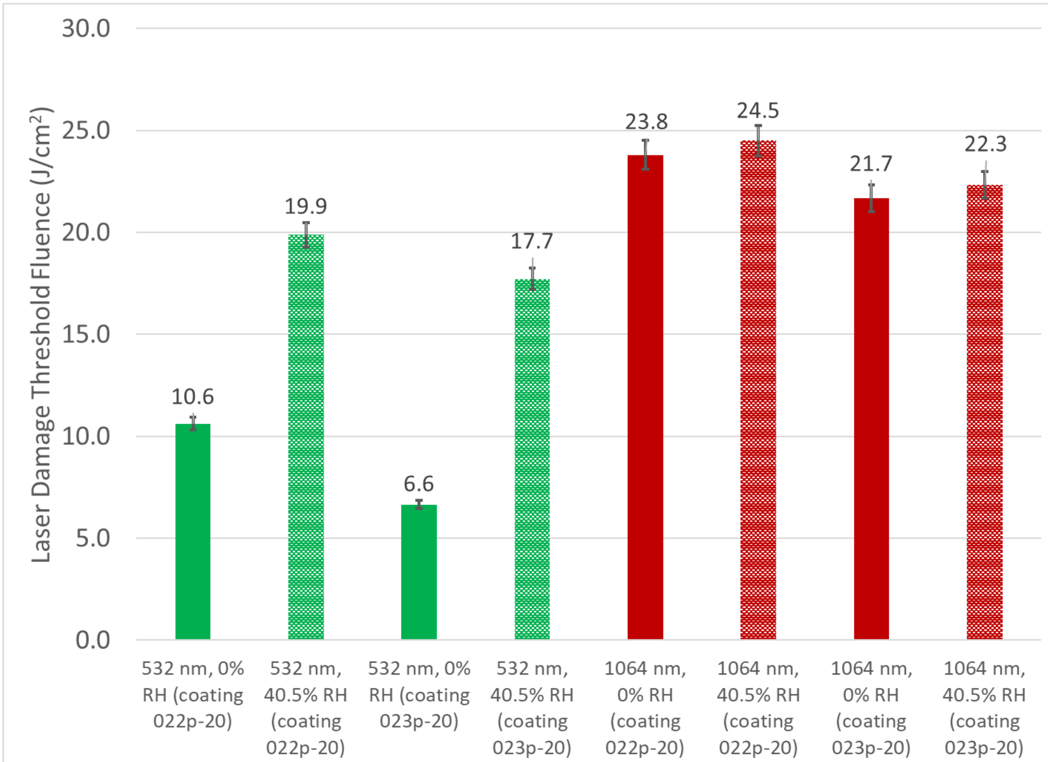

Figure 9. LIDT results of both antireflection coatings at 532 nm and 1064 nm, and 0% and 40.5% RH, at 3.5 ns pulse width and normal incidence.

Figure 10 shows the number of non-propagating damage sites that occurred during the laser damage tests. At 532 nm, the humidity delayed the onset of damage, which resulted in higher LIDTs at 40.5% RH compared to 0% RH. In other words, it seems that humidity mitigated the lower fluence precursors at 532 nm. The same effect was not seen in the 1064 nm tests perhaps because there were fewer low-fluence precursors due to the lower photon energy at 1064 nm.

These results indicate that humidity can improve the damage behavior of defects at lower fluence levels, but whether humidity can improve intrinsic damage thresholds of the thin film remains a topic for future study. Furthermore, because a dry nitrogen environment was used for the LIDT tests at 0% RH, it would be worth examining in future work to understand whether dry nitrogen is an appropriate surrogate for dry air. Based on the results of this study, there is some evidence that the substitution was appropriate because the 1064 nm LIDT results are nearly the same for 0% and 40.5% RH. However, this topic deserves further investigation.

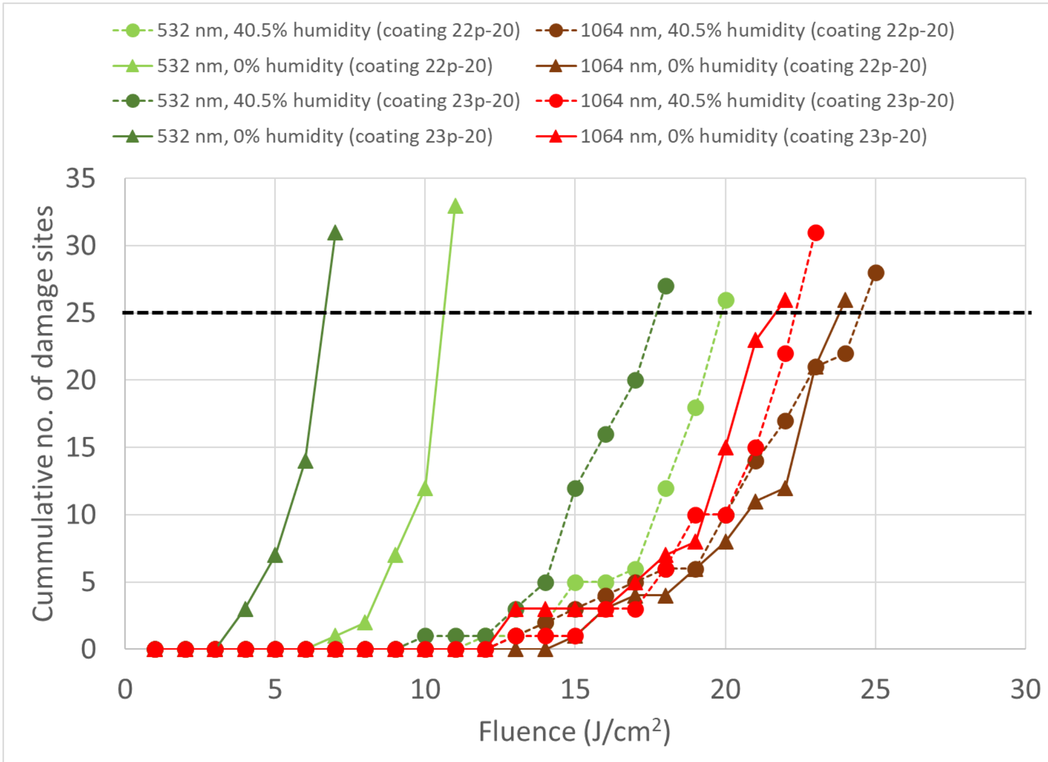

Figure 10. The cumulative number of damage sites observed during the LIDT tests of both antireflection coatings at 532 nm and 1064 nm, and 0% and 40.5% RH, at 3.5 ns pulse width. These results show that humidity delayed the onset of damage at 532 nm, thereby mitigating the lower fluence precursors at this wavelength.

4. DISCUSSION

To understand how humidity mitigated low-fluence damage precursors at 532 nm, many factors have been considered based on a review of the literature, including how humidity may affect energy transport, electric field distribution, coating stress, and surface cracking. Each of these topics are discussed in more detail below with explanations of how they relate to the LIDT results from this study. It is also of note that these effects can be intertwined.

4.1 Humidity could modify how energy couples into and away from defects and coating layers

The damage caused by nanosecond laser interactions occurs on a time scale that is long enough to involve thermal effects [15-17]. The presence of water in a porous coating could influence how light couples to defects and coating layers, possibly through bulk changes to the refractive index or by providing smoother optical interfaces/transitions around micro- or nano-scale defect sites. Also, water may aid in conducting heat away from defects sites that have been heated. There is also evidence in the literature that indicates nano-scale topology can influence the amplitudes and distributions for the density of states for allowed phonon waves [18]. In other words, the presence of water could open phonon pathways to mediate faster heat dissipation.

This is essentially a suggestion that water vapor improves the heat capacity and transport of energy from the laser spot, or it affects phonon coupling of energy transport from the laser spot. Either way, the hypothesis is that humidity allows energy to couple out of the focal spot region during the time frame of laser damage, thus improving the LIDT. This hypothesis could be tested using a shorter pulse width for laser damage testing, which would reduce the opportunity for such outcoupling of energy to occur and the LIDT would therefore be similar with and without humidity.

Jensen et al conducted LIDT testing in vacuum and ambient air on high power laser optics [3]. They attribute differences in performance to water adsorption, in part because the low-porosity coatings created with ion beam sputtering (IBS) and ion assisted deposition (IAD) showed no LIDT difference between air and vacuum, while the porous e-beam coatings did.

Also of interest is that their results from LIDT tests conducted at 355 nm showed a large difference between air and vacuum, while the 1064 nm testing did not show the same pronounced difference. This is similar behavior to what we have observed in this study because we also did not see pronounced LIDT differences at 1064 nm for 0% and 40.5% RH. Furthermore, Jensen et al showed the morphology differences between air and vacuum for an e-beam coating damaged at 1064 nm [3]. While visibly different, we considered that the damage morphology in vacuum looked tighter, possibly indicating better energy localization. This may suggest that the energy outcoupling hypothesis was at play, and supports the need for obtaining our own images of damage morphology for further analysis.

4.2 Humidity could shift the electric field distribution to a lower peak field

The incorporation of water in a porous coating is known to change the optical thickness of the coating layers [5, 19]. Therefore, humidity can cause a porous coating to experience a large red shift due the higher refractive index of water relative to air in the coating layers. However, the relatively thin AR coatings tested in this study experienced a spectral shift of only about 4 nm due to humidity, which was not enough to cause significant changes in spectral performance or the electric field distribution. In other words, the impact of humidity-induced spectral shift on electric field enhancement seems negligible for the AR coatings that we tested. However, this electric field impact could be quite different and more significant for a thicker multilayer stack or a coating with high sensitivity to spectral shift, for example, narrowband filters and mirrors.

4.3 Humidity could affect the coating stress advantageously

Studies have alluded to the impact of humidity on coating stress [4, 5]. Specifically, Anzellotti et al reported that the addition of humidity increased compressive stress in the coating [5]. Their results indicated that their coating made the transition from tensile to compressive stress at around 45% RH, which is close to the 40.5% RH that was used for LIDT testing in this study. In this scenario, we could hypothesize that if a stressed coating has a lower LIDT, then adding humidity to reduce the stress may improve the LIDT. In future work, it would be informative to measure both coating stress and LIDTs at incremental levels of humidity between the 0% and 40% RH levels. However, because our LIDT results in this study at 1064 nm were nearly the same at 0% and 40.5% RH, the contribution of coating stress is not clear.

4.4 In conjunction with reducing stresses, humidity could mitigate the presence of micro-cracking on the coating surface

At Laser Megajoule, Lavastre et al reported that polarizer coatings on silica substrates experienced a high density of cracking as soon as these components were placed in a dry environment (RH < 2%) [20]. If the surface topology (roughness, micro-cracking, etc.) is different with humidity versus without, we expect the LIDT to be impacted as well. This hypothesis is based on findings that microscopic cracks lead to localized electric field enhancement [21]. Also, the thermal conductivity of a thin film is dependent on the structure and surface qualities of the film [22]. At this point, it is unknown whether the relatively thin AR coatings tested in this study also experienced cracks or other surface flaws due to lack of humidity. However, considering that only the LIDTs at 532 nm and not 1064 nm were impacted by the humid environment, this might suggest that the surfaces of our AR coatings remained relatively intact in the dry environment. Therefore, we do not expect surface cracking to play a strong role in our LIDT results, but more investigation would be required to confirm this.

4.5 Summary

Future studies should account for energy transport and coating stress, which will lead to better understanding of how humidity mitigated the low-fluence damage precursors at 532 nm. Specifically, the coatings could be tested at shorter pulse widths and the damage morphology images could be analyzed to estimate the role of humidity in the coupling of laser energy to the coating. Also, measuring coating stress and LIDTs at different humidity levels would provide insight into the relationship between humidity-induced coating stress and LIDT.

5. CONCLUSIONS

Porous antireflection coatings for operation at both 532 nm and 1064 nm were subjected to 0% and 40.5% relative humidity environments and were tested for laser damage at 3.5 ns and normal incidence. At 1064 nm, there was no notable improvement in laser damage resistance between the dry and humid conditions. However, at 532 nm, humidity resulted in improved laser damage resistance by a factor of about two or three. Specifically, humidity delayed the onset of damage at 532 nm, effectively mitigating low-fluence damage precursors. This suggests that these coatings could be used in a humid environment to improve their durability and lifecycle in high fluence laser applications. The reduction of low-fluence precursors in a humid environment may also have implications for laser conditioning, since laser conditioning in a humid environment would allow low fluence precursors to dissipate without leading to damage spots in the coating.

This was a preliminary study and therefore the mechanisms underlying these results require further investigation. The effects of humidity on energy transport and coating stress should be addressed in future work, as there is strong evidence from previous studies indicating the impact of humidity on the physical integrity of coatings, as well as outcoupling of the laser energy from the coatings, thereby minimizing electric field enhancement at lower fluence damage precursors.

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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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