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Abstract. We have examined how three different cleaning processes affect the laser-induced damage threshold (LIDT) of antireflection coatings for large dimension, Z-Backlighter laser optics at Sandia National Laboratories. Laser damage thresholds were measured after the coatings were created, and again 4 months later to determine which cleaning processes were most effective. Coatings that received cleaning exhibited the highest LIDTs compared to coatings that were not cleaned. In some cases, there is nearly a twofold increase in the LIDT between the cleaned and uncleaned coatings (19.4 J/cm² compared to 39.1 J/cm²). Higher LIDTs were realized after 4 months of aging. The most effective cleaning process involved washing the coated surface with mild detergent, and then soaking the optic in a mixture of ethyl alcohol and deionized water. Also, the laser damage results indicate that the presence of nonpropagating (NP) damage sites dominates the LIDTs of almost every optic, despite the cleaning process used. NP damage sites can be attributed to defects such as nodules in the coating or surface contamination, which suggests that pursuing further improvements to the deposition or cleaning processes are worthwhile to achieve even higher LIDTs. © The Authors. Published by SPIE under a Creative Commons Attribution 3.0 Unported License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI. [DOI: [10.1117/1.OE.53.12.122516](https://doi.org/10.1117/1.OE.53.12.122516)]

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1 Introduction

The cleaning of optically polished substrates for optical coating purposes commands significant interest from scientists and engineers dealing with optics and lasers, as is evident from scholarly treatises¹⁻⁵ that address this topic in chapters or special sections. The optics industry also places importance on optics cleaning.⁶ These sources convey that common cleaning methods for optics include wiping the optic with a solvent such as alcohol or acetone, or immersing the optic in an ultrasonic bath that could include many steps of washing and rinsing with neutral detergents. Other cleaning methods involve the use of acids or piranha. Because these chemicals are known to attack optical coatings, they are most appropriate for cleaning more durable, uncoated substrate surfaces and should be used only with extreme care on coated surfaces so as not to damage the coatings.

These published sources of information¹⁻⁶ are very helpful and informative but, along with presenting standard cleaning protocols and precautions, they all emphasize that optics cleaning is both a science and an art, and an optic or coating is “clean” as long as it is good enough (a) for subsequent processing or (b) to ensure its reliability for its intended use. In short, the cleaning of optics and coatings is multifaceted, as are their applications, and sorting through standard protocols to arrive at effective cleaning methods for specific optics and purposes is very much a trial-and-error process governed by whether the optics or coatings meet the specifications of their particular uses

or next process steps. Methods that work for one purpose do not necessarily work for another, and the practice of holding as proprietary the methods that do work is quite understandable.

We are responsible for producing optical coatings for the large, meter size optics of Sandia’s Z-Backlighter lasers,⁷ which are linked to the most powerful and energetic x-ray source in the world, the Z-Accelerator.⁸ These lasers are kilojoule class systems providing terawatt and petawatt pulses with nanosecond pulse lengths at 527 nm, and nanosecond to subpicosecond pulse lengths at 1054 nm. Of utmost importance is that the coatings, whether for antireflection (AR) or high reflection (HR), have a laser-induced damage threshold (LIDT) that is high enough to handle the petawatt-level fluences in the Z-Backlighter laser beam trains.⁹ These two aspects, meter size substrates and high LIDT, pose a challenge in determining the applicability of standard optics cleaning protocols. Many protocols work well for small optics but are impractical for large optics. In addition, the extensive literature on optics cleaning provides surprisingly little information or data on the relationship between LIDT and standard cleaning protocols or specific cleaning methods.

Finding methods of cleaning both uncoated and coated surfaces of meter-scale optically polished substrates in a way that minimizes the prospects of laser damage are important to our coating operation. This means cleaning methods that remove as much as possible the extrinsic nanoscale defects, such as particulates, residue, and contamination, which can absorb laser energy and initiate laser damage.¹⁰ Cleaning cannot remove other extrinsic defects such as

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ejected particulates from electron beam evaporation of coating materials, and cannot remove intrinsic defects such as scratches and digs on a substrate surface due to optical polishing, or microstructural abnormalities in a coating arising from thin film growth anomalies.¹⁰

Particulates, residue, and contamination adhere to a surface or coating from the environment or during processing. For example, airborne particulates can be attracted electrostatically to an uncoated substrate surface prior to being coated, and wind up being embedded as nodules in the coating.¹⁰ We mitigate this source of defects by handling the large optics in a class 100 clean room environment. Contamination for an uncoated substrate could be trace levels of polishing compound embedded in the surface microstructure or within the redeposition layer (Beilby layer) resulting from the optical polishing process.¹¹ We confirmed the presence of such polishing compound contamination in an earlier study¹² of LIDTs of our standard 527 nm/1054 nm dual wavelength, HfO₂/SiO₂ AR coatings under exposure to nanosecond laser pulses at 1064 and 532 nm, and to subpicosecond laser pulses at 1054 nm.

Contamination for a coating could come from hydrocarbons migrating into the coating from a vacuum or ambient environment. Jitsuno et al.¹³ and Murakami et al.¹⁴ have reported on the relationship between LIDT and methods of cleaning this type of contamination from mirror coatings. Cleaning that improves LIDT by removing particulates, residue, and contamination also ensures that the substrate surface is ready to be coated. Furthermore, coating molecules adhere more effectively to an optically polished surface when it is free of particulates, residue, and contamination.

Defects of the substrate surface and the coating tend to play comparable roles for thin AR coatings while the defects within the coating and its layer boundaries play a stronger role than those of the substrate surface for multilayer HR coatings.¹¹ The reason both coating and substrate surface defects have comparable roles in laser damage for thin AR coatings is that these coatings by design allow ~99% transmission of incident laser light, so nearly the full intensity of an incident laser pulse penetrates through the coating layers to the substrate. Thus, the defects of both the coating as well as the substrate surface are exposed to the intense laser pulse and can comparably initiate laser damage.^{11,15} We have presented examples of such electric field behaviors of our AR coating designs in previous papers.^{16,17} These AR coatings consist of 2 HfO₂/SiO₂ layer pairs with a total thickness of ~0.5 μm.

AR coatings are of special importance to us because a major responsibility of Sandia's large optics coating operation^{9,18} is to provide AR coatings on both sides of Z-Backlighter debris shield windows, which are flat, fused silica substrates measuring 32.5 cm × 32.5 cm × 1 cm. They are used to protect other Z-Backlighter laser optics from high-velocity projectiles released by experiments conducted in the Z-Accelerator. Each Z experiment releases enough debris to cloud the surface of a debris shield, which means it cannot be used for more than one Z experiment. Nevertheless, the debris shield AR coatings must have LIDTs of at least 20 J/cm² so that the high intensity Z-Backlighter laser pulse can pass through them without causing damage that would prevent the pulse from arriving to focus at its full fluence. Every year, we provide AR

coatings for approximately 50 debris shields, in addition to AR coatings for numerous other meter-class Z-Backlighter lenses and windows.

We conducted a study of the relationship between cleaning methods and LIDT for AR coatings even though the production demands for coatings on large optics afford us limited time for research investigations. For this reason, we included only four test optics in the study and coated them in regular large optics coating runs. Our study specifically focuses on how LIDT depends on the protocols for cleaning an already AR coated optic as opposed to protocols for preparing an uncoated substrate surface for coating. We chose to vary the protocols for cleaning already AR coated optics because this cleaning process presents more challenges to us while the protocol we use for cleaning uncoated surfaces is, in our experience, well established and reliable.^{9,12}

Sections 2 and 3 describe the cleaning protocols for our tests and our rationale for choosing them, and then Secs. 4 and 5 present the experimental procedure used and the results obtained. Section 6 gives our conclusions.

2 Cleaning Protocols

One of the challenges just mentioned regarding cleaning of AR coated substrate surfaces that is common to all optics for which both sides of the substrate need to be coated has to do with one side of the substrate being coated first and the opposite, uncoated side still needing to be coated. In this situation, the first coated surface is a "ride along" not only during the process of cleaning the uncoated side but also for the process of coating it. These cleaning and coating processes for the uncoated side can expose the first coated side to particulates, residue, or contamination. In cycling back through the coating run for the uncoated surface of the optic, its first coated surface faces up in the coating chamber. This exposes the already coated surface to possible particulate and bearing grease contamination that can fall down on it from rotating planetary gears and coating chamber ceiling panels. Complete elimination of this type of contamination is difficult to achieve but it can be effectively reduced by use of low out-gassing Krytox grease for the bearings, blocking pathways for contamination to descend on an up-facing optic surface, and periodically cleaning the wall and ceiling panels of the chamber. An additional aspect of "ride-along" coatings is that depositions often occur at a chamber temperature of 200°C, which requires that large optics be thermalized at this temperature overnight, so the up-facing coating undergoes many hours of thermal conditioning. In our experience, this thermal conditioning allows the coating to hold particulates, residue, or contamination more strongly. In general, almost every time an up-facing coated surface "rides along" through a coating run, this coated surface is usually much more difficult to clean than the coated surface that has not cycled back through a coating process as a "ride along."

Cleaning of all of our large optical substrate surfaces, whether uncoated or coated, is performed by hand using mechanical scrubbing with ultralow particulate, hydro-entangled polyester/cellulose Texwipes in conjunction with generous amounts of highly deionized (DI) water (resistivity >17.5 MΩ) flowing over the surfaces. Such mechanical scrubbing force is required to overpower the adhesion forces holding nanoscale particulates, residue, and

contamination on uncoated or coated optically polished surfaces. Our bare substrates are optical glasses with high Knoop hardness, and can be scrubbed very vigorously and repeatedly using fine particulate abrasive slurries without adding to the scratches or digs of the surfaces. This vigorous abrasive scrubbing is reliably effective in removing nanoscale particulates, residue, and contamination, including polishing compound embedded in the surface microstructure.¹² Our coatings, on the other hand, while consisting of hard dielectric oxides, are amorphous, with porous, columnar structure, making their surfaces not nearly as hard as bare substrate surfaces. So, the use of gentler scrubbing without abrasive slurries is required for coated surfaces even though more vigorous and abrasive scrubbing might remove nanoscale contamination as effectively as it does for bare substrates. It would, however, not lead to higher LIDT because it would be removing extrinsic defects at the expense of adding scratches and digs to the surface of the coating. Accordingly, we use a mild detergent (Micro 90) as well as a fine particulate alumina slurry (Baikalox) in our standard wash protocol for uncoated surfaces, and use only the mild detergent without the alumina slurry in our standard wash protocol for coated surfaces. This gentler protocol for coated surfaces makes cleaning them very challenging, especially when they have the more extensive extrinsic contamination after a “ride along” back through a coating run.

Because our standard wash protocol for coated surfaces is not as effective as we would like, and is also quite labor intensive, we wanted to explore other cleaning methods to determine whether our existing method is still the best for upholding high LIDTs for our AR coatings. In searching the literature for alternative options, we found only a few reports^{9,13,14,18–20} on optics cleaning methods relating to LIDT, particularly for large, high power laser optics, even though the negative impact of contamination on LIDTs as a result of poor cleaning is a widely recognized problem.⁵ We also could find no industry standards for optics cleaning, and managed to identify only a limited selection of other cleaning processes to choose from and compare to our own. The option we found that held the best promise to complement and improve our current protocol for cleaning coated surfaces is the process developed by Jitsuno et al.¹³ and Murakami et al.¹⁴ It involves soaking the optic in a mixture of alcohol and DI water for a period of time to remove contamination, was developed specifically for high power laser applications, and was successful at removing oil contamination from coated optics, allowing the LIDTs of some optics to nearly double. Given the proven track record of this cleaning process,^{13,14} and the reduced amount of manual labor it involves, we decided to test it and compare it to the effectiveness of our existing cleaning method for coated optics. The following section describes in more detail our existing cleaning protocols for uncoated and coated surfaces, the method of Jitsuno et al.¹³ and Murakami et al.¹⁴ for coated surfaces, and also a hybrid protocol for cleaning coated surfaces. A subsequent section explains the setup of the LIDT tests that allow comparisons, in terms LIDTs, of the effectiveness of each cleaning method for the coated surfaces amongst themselves and against simply not cleaning the coated surfaces at all.

3 Experimental Procedure

As mentioned in Sec. 1, the focus of this study is on choosing an appropriate cleaning method to perform on a large optic after it has received an AR coating, with the goal of achieving the highest LIDT for the coated surface. We identify this postcoating cleaning as “final cleaning.” However, the first part of this section reviews and explains our standard protocol for cleaning uncoated surfaces. We have presented this protocol before,^{9,12} but not in its full context that accounts for the ride along of coated surfaces with uncoated surfaces. This information allows anyone to repeat our cleaning experiments, and allows us to disclose the processes that a large optic undergoes before final cleaning takes place.

Our standard AR optic cleaning process involves two steps: the first to clean the completely uncoated optic for coating of its first side, and the second to prepare the optic for AR coating of its second side after its first side has been AR coated. These two cleaning steps are then followed by the final cleaning of the optic after it has been AR coated on both sides, as summarized below. Each step occurs in a class 100 clean room.

Cleaning step 1: We clean each side of the uncoated optic by first rinsing with DI water while lightly wiping with a wiper (Texwipe model # TX1109) to remove large particles. Then, we vigorously wash each uncoated side of the optic with a DI water soaked wiper and mild detergent (Micro 90, which is diluted to a ratio of roughly 10:1 by volume of DI water to detergent). Micro 90 removes organic, oily residue on an optic. After rinsing away the Micro 90, we then vigorously wash both uncoated sides with a DI water soaked wiper and Baikalex. Baikalex is the slurry of $<0.05 \mu\text{m}$ alumina particles that can remove particulates and polishing compound left behind in the optical surface microstructure from the glass polishing process. We then rinse the optic with DI water and vigorously wash both sides of the optic again with Micro 90 and a DI water soaked wiper to remove any Baikalex that did not rinse away. The final step is to scrub the optic vigorously using a wiper with copious flow of just DI water to help remove any other particles and residues that remain. The optic is set to dry under laminar air flow provided by the clean room’s HEPA filtered air handlers. We repeat this cleaning step as often as needed until a visual inspection of the optic reveals that the clear aperture is free from contaminants such as dust particles and residue. The visual inspection, which is done in ambient darkness using a bright, white light, fiber optic illumination source, is part of the art of optics cleaning. After cleaning step 1, the uncoated optic is ready to receive the AR coating on its first side.

Cleaning step 2: Same as cleaning step 1, but Baikalex is not used on the first side that was coated because it can mar that coating. After cleaning step 2, the optic is ready to be coated on the second side.

Final cleaning step: Same as cleaning step 1, but Baikalex is not used at all because both sides of the optic have been coated. After this point, the optic is ready for final visual inspection and installation in the Z-Backlighter beam train.

In our experience, freshly polished and coated optics usually clean easily and do not require repeated cleanings. The same applies to this study: none of the cleaning steps were repeated on our test optics. However, optics that have been stored in dirty containers or coated in an oil-contaminated

vacuum chamber may require a lot of attention before they finally appear free of contamination. Our worst case involved a lens with an AR coating that became contaminated with oil during the coating process and subsequently underwent final cleaning 16 times before the contamination was removed from the clear aperture.

Having explained our existing final cleaning process, we now describe the final cleaning process, based on the approach of Jitsuno et al.¹³ and Murakami et al.,¹⁴ that we included in our tests. In this process, the optic is submerged for 6 h in a bath that contains equal parts by volume of DI water and alcohol. The DI water migrates into the microstructure of the coated optic surface more readily than does the alcohol, and in this way tends to displace contaminants, especially organics such as hydrocarbons. The alcohol then tends to prevent the displaced contaminants from remaining on the optic surface as a residue. After the 6-h soaking period, the optic is rinsed with DI water and then set to dry in air.

We tested, altogether, four final cleaning processes in this study, and list them below as processes A–D. Process A is our existing final cleaning method, process B is a control process that skips final cleaning altogether, and processes C and D are new and involve the cleaning method of Jitsuno et al.¹³ and Murakami et al.¹⁴ We used four 50-mm diameter, 10-mm thick fused silica test optics, one for each of the four final cleaning processes, A–D. The test substrates were double-sided polished using ceria polishing compound. We chose for the tests our standard AR coating for a normal angle of incidence at 1054 nm.¹⁸ It consists of 2 HfO₂/SiO₂ layer pairs, with a total thickness of ~0.44 μm. The four test optics were coated with the AR coating on both sides, in the same production coating runs in Sandia's large optics coating chamber, with deposition via e-beam evaporation. SiO₂ was evaporated at a rate of 7 Å/s. Hf was evaporated at a rate of 1 Å/s for the first layer and 2 Å/s for the second layer, in a reactive process with O₂ gas, resulting in a pressure of 1.2×10^{-4} Torr in the coating chamber. The deposition occurred at 200°C. The AR coatings deposited for side 1 and side 2 are nearly identical, as shown by the transmission scan in Fig. 1, which was captured by the Perkin-Elmer Lambda 950 spectrophotometer. The side 1 coating was

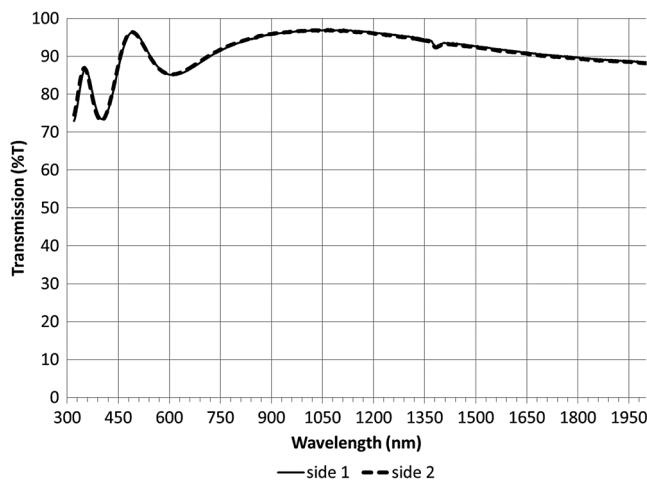


Fig. 1 Transmission scans at normal incidence of the antireflection (AR) coating that was deposited on side 1 and side 2 of the test optics. The transmission scans are nearly identical.

deposited on December 11, 2013 and the side 2 coating was deposited on December 13, 2013.

All of the test optics first underwent cleaning steps 1 and 2 before undergoing the final cleaning, according to one of four final cleaning processes as listed below:

- A. Perform our existing final cleaning method.
- B. Skip final cleaning entirely. (The optic is alcohol-wiped before LIDT testing by the testing company).
- C. Perform final cleaning based on the method of Jitsuno et al.¹³ and Murakami et al.:¹⁴ soak the optic in a DI water/ethyl alcohol (1:1 by volume) bath for 6 h, and then rinse it in DI water for 30 s.
- D. Same as process C, followed by our existing final cleaning method.

4 LIDT Tests

Each optic, after final cleaning according to one of the four processes, A–D, described above, underwent LIDT testing in late December 2013, on both sides at 1064 nm and normal incidence according to the NIF-MEL protocol.²¹ To determine whether the final cleaning procedures have an effect on the LIDT of the AR coatings as they age, we performed the final cleaning procedures on the optics again after a period of 4 months and had the LIDTs remeasured in April 2014. Another reason that we wanted to repeat the cleaning tests after 4 months is that we usually clean optics again before they are installed in the beam train if they have been in storage for several months. Although the optics were in storage for 4 months, they were housed in clean containers made from polyethylene terephthalate glycol-modified (PETG).

The laser damage tests were performed according to the NIF-MEL test protocol by Spica Technologies, Inc.²² In this protocol, after the coated surface undergoes an alcohol drag-wipe cleaning step, 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 cm² 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/cm² in the cross section of the laser beam. After testing the 2500 sites at 1 J/cm², the fluence is increased in a 3 J/cm² increment and the 2500 sites are tested again, and this progression repeats until the damage threshold fluence is reached. For our AR coatings, as we explained earlier, the intrinsic and extrinsic defects of both the substrate and the coating play comparable roles in initiating laser damage. This will help us to understand the LIDT results that we obtained.

The NIF-MEL procedure is essentially an N-on-1 test at each of the 2500 sites. Laser damage will be some type of melt or crater that alters the AR 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. In other cases, the damage does propagate. The LIDT is reached at the fluence at which 1 or more propagating damage sites occurs, or the fluence at which the number of nonpropagating (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 cm² coating area tested. Our reasoning behind this LIDT criterion is the following. We know we cannot tolerate a propagating damage site in the Z-Backlighter laser beam trains 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 cm², 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.

An important aspect of the NIF-MEL protocol is that it probes a coating for laser damage in a statistically significant way and in a way that is not just at widely separated sites. The 2500 sites are dense over the 1 cm² area, with about 200 μm from one site to the other in the raster scan, and the 90% radius of the peak intensity of the 1-mm diameter beam is 100 μm. This means that laser pulses thoroughly expose the entire 1 cm² area of the coated surface in each raster scan, increasing the probability that the random distribution of nanoscale intrinsic and extrinsic defects over the substrate and coating surfaces are accounted for in the laser damage tests. This is in contrast to testing at widely separated sites, which may miss important defect zones and thus provide false assessment of a coating's LIDT. The multilongitudinal mode nature of the laser pulses also provides a statistical averaging effect in the NIF-MEL tests. Such nanosecond pulses exhibit random intensity fluctuations and spiking on the ps scale. Only the total pulse energy is measured in the tests, and then converted into average fluence for the pulse, so the actual fluences to which each test site is exposed are a random distribution about the average fluence. This random spiking in time at the ps scale of the NIF-MEL laser pulses has a counterpart in the Z-Backlighter pulses in the form of random intensity hot spots in space over the ~1000 cm² beam cross section. This means the NIF-MEL protocol tests for LIDT under conditions that are, in a statistical way, somewhat similar to the conditions for laser damage to large optics in the Z-Backlighter beam train.

The instrument for the NIF-MEL average fluence measurements has an absolute accuracy of ±3%.²³ For the LIDT based on accumulation of NP damage sites, we identify the highest test fluence for which fewer than 25 damage sites have accumulated. This means the next test fluence, which is 3 J/cm² higher in the NIF-MEL fluence progression, is the lowest test fluence for which >25 damage sites have accumulated. Using the ±3% accuracy of the fluences, we interpolate linearly between these two fluences to establish the range of fluences for which at least 25 NP damage sites would occur consistent with the data for the NIF-MEL test fluences. If the accumulation of NP damage sites happens to be exactly 25 at a NIF-MEL test fluence, then we take the ±3% accuracy for that test fluence as the range of fluences for which at least 25 NP damage sites occur. These ranges of fluences establish the measurement uncertainties for our reported LIDTs on the basis of accumulation of NP damage. For the LIDT based on one or more propagating damage sites, we also take the ±3% accuracy for the NIF-MEL test fluence at which propagation was detected as the measurement uncertainty for the LIDT based on propagating damage.

5 Results

The LIDT results for each optic are shown in Fig. 2 for the LIDT tests that were performed in December 2013 and April 2014. Taking the average of the side 1 and side 2 LIDTs, the December 2013 averages are 33.5, 21.0, 30.3, and 30.8 J/cm² for processes A, B, C, and D, respectively. The April 2014 averages are higher than those from December 2013, and they are 38.0, 29.6, 38.5, and 32.8 J/cm² for processes A, B, C, and D, respectively. The LIDTs are highest on average for final cleaning process A (our existing final cleaning process), and processes C and D that involve the DI water/alcohol soak. For cleaning process B that skips the final cleaning process altogether, on the other hand, the LIDTs are lowest on average, and the side 1 LIDT is below our required LIDT of 20 J/cm². This confirms the importance of the final cleaning in achieving the highest LIDTs, and indicates that the DI water/alcohol soak can be effective as a final cleaning option.

The LIDTs differ between side 1 and side 2 of the same optic. We are not surprised by this when considering that the uncoated sides of the test optics in our experiment may differ in their nanoscale defects due to polishing variations

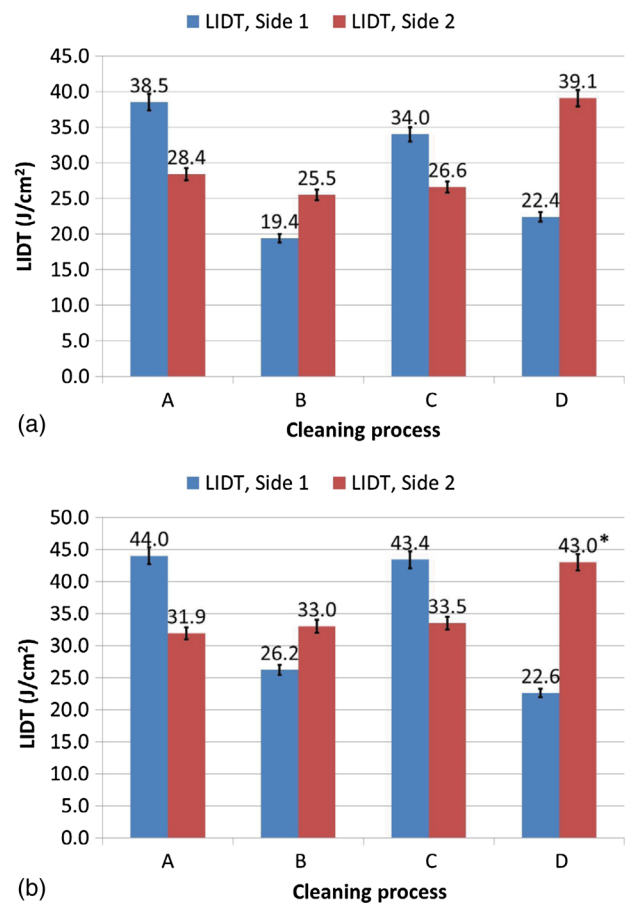


Fig. 2 Laser-induced damage thresholds (LIDTs) at 1064 nm and normal incidence for side 1 and side 2 of each AR coated test optic with respect to the cleaning process used. These LIDT measurements were taken in December 2013 (a) and April 2014 (b). The asterisk in (b) indicates that the maximum fluence on side 2 for cleaning process D corresponds to only 16 NP sites, but the LIDT test could not be advanced further as a result of propagating damage detected on the substrate of side 1.

that can occur even though they were all polished similarly. Since LIDTs of AR coatings are influenced comparably by coating and substrate defects, we expect such defect differences of the underlying test optic surfaces to be reflected in the LIDTs. In all cases, the side 1 coating receives a mild detergent wash (cleaning step 2) in between the first and second coating runs, but then undergoes the rather harsh conditions of facing up in the chamber at 200°C as a ride along during the side 2 coating. As explained earlier, fine particulate contamination from the planetary fixture drive might fall down on side 1, and the coating simply bakes at 200°C for the roughly 12 h the chamber is at that temperature prior to and during the side 2 coating run. For these reasons, if no cleaning occurs after both sides are coated, we expect the first side coated (side 1 in this study) to be more susceptible to laser damage than the second side coated (side 2 in this study). The LIDT results for process B (no final cleaning) confirm this. The LIDT results for processes A and C indicate, on the other hand, that the first coated side, with final cleaning, suffers no degradation of laser damage resistance due to having been in the coating chamber facing up during the coating of the second side.

The LIDT results for final cleaning process D show LIDTs for side 1 that are significantly less than for side 2, and that are also significantly less than for the other side 1 coatings except for process B, for which there was no final cleaning of the coatings at all. The low LIDT of side 1 for cleaning process D can be attributed to defects on the surface of the substrate. Although the LIDT testing company was performing the damage tests on side 2 of the optic in April 2014, propagating damage appeared on the substrate of side 1. The testing company therefore stopped performing the damage test according to NIF-MEL test protocol, which means that the NP damage threshold on side 2 of this optic is actually higher than what we have reported in the April 2014 data. Furthermore, we do not expect that this low side 1 LIDT for process D is due to defects of the coating because all side 1 coatings are from the same coating run. Also, process D final cleaning is at least as thorough as process A or process C final cleaning since it is a combination of both processes A and C. This indicates that the low side 1 LIDT for process D can be explained by defects of the underlying substrate surface and does not imply a deficiency of process D as a final cleaning process.

The LIDT of each coating increased after a period of 4 months. The amount that each LIDT increased is shown in Fig. 3. The coating cleaned by process C exhibited the greatest LIDT increase, but this result does not completely reflect the effect of cleaning because the optic that did not undergo final cleaning (i.e., cleaning process B) also displayed increased LIDTs. This indicates that an aging effect in the coatings may have contributed to the increased LIDTs. Damage test conditions could also play a role. Aging effects are common, and can result in wavelength shifts in the transmission spectrum of multilayer HR coatings,²⁴ but here, in the case of this four-layer AR coating, such wavelength shifts were not evident, as was confirmed by a remeasurement of the transmission spectrum of each coating again in the spectrophotometer. Nevertheless, aging resulting from alterations in molecular bonding within the coating structure could lead to relaxation of stress around voids of the

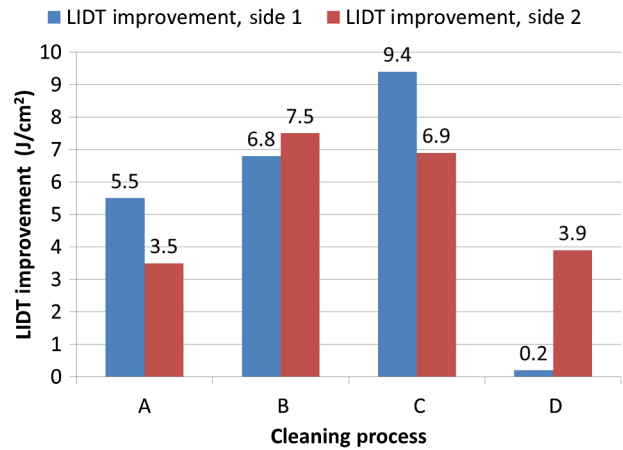


Fig. 3 The amount that the LIDT increased after each coated optic underwent final cleaning again, after being stored in a clean polyethylene terephthalate glycol-modified (PETG) container for 4 months.

columnar coating formations and cause the coating to become more resistant to laser damage. Further studies into aging, for example, by taking stress measurements and repeating the cleaning experiments after leaving the optics in a dirty container for >1 year, may provide a more accurate assessment of the effectiveness of each cleaning process in removing contamination.

It is interesting that the LIDT for side 1 of the optic that underwent cleaning process D exhibits almost no aging effect. This is consistent with our assessment above that this side 1 laser damage is primarily due to defects of the underlying substrate surface, and these defects, being independent of the coating or its aging, governed the laser damage the same in December 2013 as they did in April 2014. This shows that, for few-layer AR coatings, the substrate and coating defects are like a double-edged sword with respect to LIDT. Both influence laser damage, but sometimes one dominates over the other in its influence. When the substrate defects dominate, no final cleaning method can lead to improved LIDT because final cleaning, of course, only can remove the extrinsic defects of the coating and not the defects of the underlying substrate surface.

Figure 4 shows the cumulative number of NP damage sites versus the laser fluence that each coating exhibited during the laser damage tests. This is the NP damage site data that we used, as we explained earlier, for determining the LIDTs of Fig. 2. NP damage sites tend to result from surface or coating defects for AR coatings, as we have explained earlier, which means that improving the quality of the polishing process or deposition process, or improving the cleaning methods even more, should result in coatings with lower defect densities and hence higher LIDTs.^{5,10,11,25,26}

The NP damage site accumulation data of Fig. 4 gives insight into how the LIDT behaviors of Fig. 2 correlate to the effectiveness of the different final cleaning processes. Figure 4(a) contains data from the LIDT tests performed in December 2013. It shows that the data for cleaning processes A (side 1) and D (side 2) behave similarly and almost overlap. These are also the two data sets which yielded the highest damage thresholds at that time. Likewise, the data for cleaning processes A, B, and C on side 2 also behave similarly and almost overlap. This demonstrates that different cleaning methods can result in similar LIDT outcomes

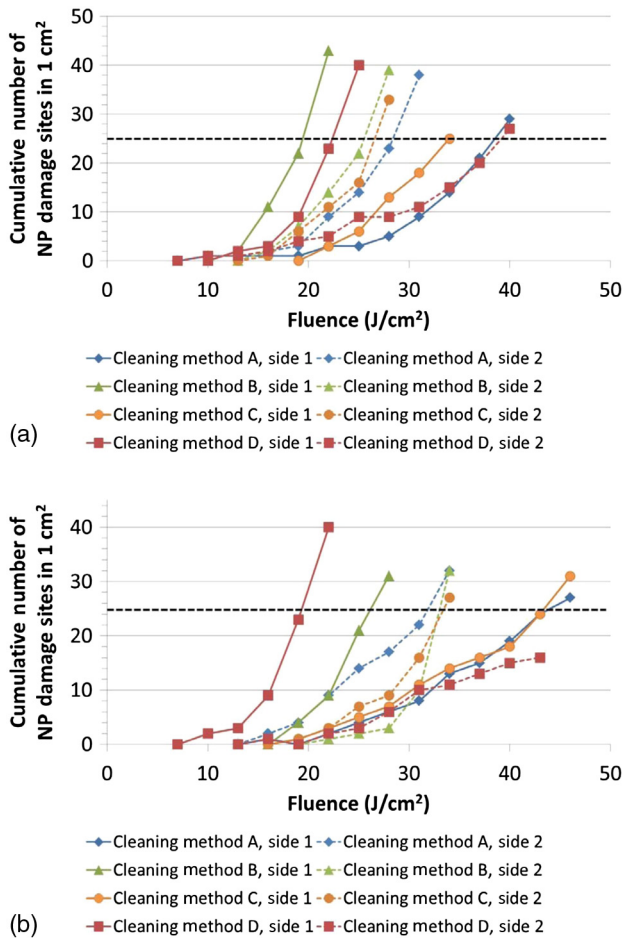


Fig. 4 The cumulative number of NP damage sites versus laser fluence results from the NIF-MEL LIDT tests performed in December 2013 (a) and April 2014 (b). When the data intersect the horizontal dashed line, this indicates the fluence at which 25 NP damage sites were observed, and hence sets the LIDT of the AR coating according to the NIF-MEL protocol.

and hence we are unable to predict from this data alone which cleaning method is best. However, given that the lowest damage thresholds on average occurred for the optic that did not receive final cleaning (cleaning process B), and that the side 1 LIDT of this optic was below our requirement of 20 J/cm^2 , we conclude that final cleaning is essential toward achieving the necessary higher LIDTs.

Figure 4(b) contains data from the LIDT tests performed in April 2014. Unlike the LIDT data from December 2013, it now shows that the data for cleaning processes A (side 1), C (side 1), and D (side 2) are similar, almost overlap, and are more linear than they were previously. The data for cleaning methods A (side 2) and C (side 2) have also become more linear. We do not attribute this more linear behavior to an aging effect in the coatings because we do not observe this behavior in the optic that received cleaning process D on side 1, or the optic that received no final cleaning (cleaning process B). Nonetheless, the linear behavior is a desired effect because it dampens the number of NP damage sites generated at lower fluences. We believe the linear behavior is connected to lower contamination levels on the surface of the coatings as a result of undergoing a second cleaning process. The fact that the data in Fig. 4(b) for side 1 of the optic

that underwent cleaning process D does not exhibit linear behavior despite being cleaned is further confirmation of our conclusion above that laser damage in this case is primarily due to intrinsic defects of the underlying optic surface. With the high levels of NP damage sites attributable to defects of the substrate surface, it appears that no amount of final cleaning could have overcome the effect of the substrate defects on the LIDT.

6 Conclusions

Each final cleaning method tested in this study resulted, on average, in higher LIDTs compared to no final cleaning of an AR coating. A special exception is the optic that underwent cleaning process D, which is a combination of our existing cleaning method involving a detergent wash, and the alcohol/DI water soaking method developed by Jitsuno et al.¹³ and Murakami et al.¹⁴ The suspected presence of intrinsic defects on side 1 of the substrate that received cleaning process D dominated the LIDT to a degree that cleaning could not overcome, and resulted in approximately 50% lower LIDTs compared to side 2 of the same optic. Therefore, the effectiveness of cleaning method D is not well represented on side 1 of this optic. This is also the case for side 2 because LIDT testing in April 2014 was terminated on side 2 due to propagating damage observed on side 1 of the substrate that can be explained by substrate defects on side 1; hence, the LIDT on side 2 is likely higher than what we have reported. This indicates that cleaning process D could have resulted in the highest LIDTs compared to the other cleaning processes that we tested, and may be the best cleaning method for achieving high LIDTs for AR coatings. This conclusion seems appropriate because cleaning process D was also the most rigorous final cleaning process that we tested, and thus likely to be most effective.

We have learned from this study that the LIDTs that can be achieved using our current final cleaning method are nearly the same as the LIDTs that can be achieved using a final cleaning method that requires fewer physical demands, which consists of soaking the optic in equal parts of DI water and ethyl alcohol for 6 h. We have used this DI water/alcohol final cleaning method for our debris shields, though for larger optics it may be cumbersome. Also, some facilities may find it problematic to manage the hazardous waste generated by the gallons of alcohol used for soaking optics. Exploring the tradeoffs between hazardous waste, time commitment, optic size, and labor intensity should, therefore, also guide the decision about which final cleaning method to implement.

Another outcome of this study was the discovery that our AR coatings may undergo an aging effect that results in higher LIDTs after a 4-month period. The enhanced LIDTs of our AR coatings after aging warrants further investigation, but in the meantime, we appreciate that allowing an AR coated optic to age for a few months may be a simple strategy to gain higher LIDTs with little effort.

Most of the optics in this study reached their LIDT as a result of NP damage sites, which we attribute to coating defects and surface contamination. Resolving problems with either the quality of the coatings (removing nodules¹⁰), or making further improvements to the cleaning method, should lead to higher LIDTs. We look forward to performing similar cleaning experiments on other coatings such as HR

coatings. We also wish to learn more about how effective these cleaning processes are on improving the LIDTs of optics that have gotten dirty as a result of being stored for more than a year in containers that are not well sealed, or in containers that outgas.

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