

Combined Advanced Finishing and UV-Laser Conditioning for Producing UV-Damage-Resistant Fused Silica Optics

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ABSTRACT

Laser induced damage initiation on fused silica optics can limit the lifetime of the components when used in high power UV laser environments. For example in inertial confinement fusion research applications, the optics can be exposed to temporal laser pulses of about 3-nsec with average fluences of 8 J/cm² and peak fluences between 12 and 15 J/cm². During the past year, we have focused on optimizing the damage performance at a wavelength of 355-nm (3 ω), 3-nsec pulse length, for optics in this category by examining a variety of finishing technologies with a challenge to improve the laser damage initiation density by at least two orders of magnitude. In this paper, we describe recent advances in improving the 3 ω damage initiation performance of laboratory-scale zirconium oxide and cerium oxide conventionally finished fused silica optics via application of processes incorporating magnetorheological finishing (MRF), wet chemical etching, and UV laser conditioning. Details of the advanced finishing procedures are described and comparisons are made between the procedures based upon large area 3 ω damage performance, polishing layer contamination, and optical subsurface damage.

Keywords: magnetic fluid, polishing abrasives, magnetorheological finishing, subsurface damage, etching, UV laser conditioning, damage threshold, surface modification, polishing layer.

1. INTRODUCTION

Methods for reducing laser damage initiation sites that lead to catastrophic failure of fused silica optics are of critical importance to the laser-driven Inertial Confinement Fusion research (ICF) community. They are also of importance to the multi-billion dollar semiconductor capital equipment market. In lithography equipment used for manufacturing of silicon integrated circuits, ultraviolet light in the range of 340-360 nm is typically used. There are many commercial lasers that use light wavelengths at 360 nm or less. The ability to substantially improve the resistance to optical deterioration of laser components in such devices is important, since the chief limiting issue for laser materials is their laser damage performance. Unfortunately, success in using conventional polishing methods has been limited in reducing the initiation of damage sites when fused silica optics are exposed to high-power lasers at these wavelengths.

The sustained performance of high peak power lasers, such as those used for inertial ICF research, is limited by the damage to optical components^{1,2} that comprise the optical chain used to deliver the laser beam to the fusion capsule. The damage initiates in small (tens of micrometers) sites whose particular size is dependent upon the peak fluence of the laser. The size of the damage grows exponentially with the number of shots and can create excessive scatter and beam modulation³. Replacing the damaged optics is an option for sustained performance of the laser system, but such an option is expensive. Methods of stopping damage growth have been recently developed, but their implementation requires a method of reducing the number of sites.

The largest and most powerful lasers designed for ICF research are flashlamp pumped neodymium-glass operate at a wavelength of 1053 nm which is then converted to a third harmonic (3 ω) at 351 nm^{4,5}. In the final optics stage of an ICF

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laser, the infrared beams are converted to ultraviolet beams, and then focused onto the target. The final optics will be irradiated by a distribution of fluences, peaking at about 15 J/cm^2 for a 3-nsec pulse at a wavelength of 351 nm. Optical components, particularly made of fused silica, are essential for shaping the laser beams and delivering these beams to the target. These optics suffer from weakened areas that are prone to damage at the laser fluences expected or planned in the aforementioned laser systems. Damage can occur at the fundamental laser wavelength of 1053 nm and, especially, at the tripled wavelength of 351 nm. There are two options that would enable continued use of the optics. One option is to mitigate the growth of the damage sites before they can create excessive scatter and beam modulation^{1,6-8}. The other option is to drastically reduce the number of initiation sites present on the optics following optical finishing⁹.

Damage initiation on fused silica optics in high fluence UV laser environments can arise from various manufacturing flaws and handling processes used during optic fabrication. Improper handling and cleanliness of the optics can cause damage initiation by introducing contaminants onto the optical surfaces. Such contaminants can absorb energy and initiate damage upon laser beam exposure. Similarly, the conventional polishing process itself can lead to the formation of damage sites¹⁰⁻¹³. Such damage sites result from both surface and subsurface mechanical damage that is inherent to the high normal loads associated with conventional lap polishing. In addition to the mechanical damage itself, such surface and subsurface cracks can serve as sites that can trap optically absorbing species, such as iron, ceria, and other contaminants, which are typically present in the polishing process. While improvement in damage initiation density can be realized by utilizing zirconia-based polishing slurries¹⁴, conventional lap polishing inevitably results in both surface and subsurface mechanical damage due to the high normal loads present from the weight of the optic on the lap bed. Because of this, the subsurface damage layer persists regardless of how careful one is in controlling the process. Similar effects are observed with other types of conventional finishing such as small tool polishing.

Magnetorheological finishing¹⁵⁻²¹ (MRF) has been recently described as an advanced polishing technique that can finish optics without propagating the subsurface damage layer. The technique can also remove preexisting subsurface damage under the correct conditions. The removal of subsurface damage using MRF can be attributed to the small normal stress applied to the glass surface compared to large shear stress which is created by the interaction of the tool's magnetic field and the MR fluid through the converging gap between the optic and tool's pole faces. The shear stress applied to the optic results in high rates of material removal with little medial crack formation.

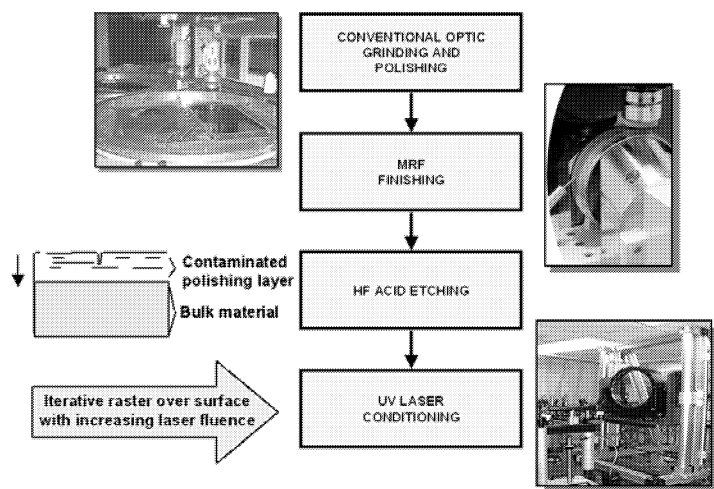


Figure 1: Combined advanced finishing recipe to improve UV damage resistance of fused silica optics.

This paper compares the results of 3ω damage testing on conventionally polished optics and optics polished using MRF. Additional post-processing procedures involving hydrofluoric acid etching²² and 3ω laser conditioning⁹ are also discussed which, when combined with conventional polishing and MRF polishing, result in improved 3ω damage resistance when compared to the use of any of the techniques when used alone. This combined advanced finishing process

provides a means of fabricating UV damage resistant optical components with minimal changes in the manufacturing process as shown in Figure 1.

2. EXPERIMENTAL

The experimental matrix shown in Table 1 is designed to explore the effects of MRF, HF etching, and UV laser conditioning on conventionally finished optics provided by different vendors. Both large aperture and subscale optics are considered in the experiments to illustrate any differences in performance due to vendor specific processes. The samples polished by Zygo consist of 15.2 cm-diameter by 1.0-cm thick fused silica optics (Corning 7980). These specimens were prepared by conventionally polishing both sides of the optics using best-known continuous pitch polishing

techniques and zirconia (ZrOx) slurry. A subset of the optics was also subjected to MRF final polishing using standard ceria (CeOx) slurry contained in an iron based MR fluid media. For these specimens, 1 μm of material was removed using MRF. The “Vendor 1”, Table 1, optics consist of subsections cut from a 43-cm X 43-cm X 2.5-cm large aperture optic (Corning 7980 fused silica) originally prepared in 2000 using a proprietary best-known process that optimizes 3ω damage performance. These optics represent our reference for the present studies since they are the best-performing conventionally finished large optics tested at LLNL. The subsections used in the experiments were square measuring 14.3-cm X 14.3-cm X 2.5-cm thick. The “Vendor 2” optics consist of subsections cut from a 43-cm X 43-cm X 1.0-cm large aperture optic originally prepared in 1999 during the early portion of our large aperture optic finishing development efforts. During the polishing of these optics, little attention was paid to the type of polishing slurry used and the extent of subsurface damage encountered during the fabrication process. The subsections were square measuring 14.3-cm X 14.3-cm X 1.0-cm thick. The LLNL-polished specimens consist of subsections cut from a large aperture optic of the same vintage as that provided by “Vendor 2”. The subsections were subjected to

Table 1: Experimental matrix used to compare various finishing and post-processing techniques on the vendor supplied optics. X identifies the processes that are explored via large area damage testing.

		As Is	Cond.	Etch	Etch + Cond.
		MRF	MRF + Cond.	MRF + Etch	MRF + Cond.
Subscale Optics	Finishing Vendor				
	Zygo ZrOx	X	X	X	X
	LLNL CeOx				X
Full-scale NIF Vendors	Vendor 1	X	X	X	X
	Vendor 2	X	X	X	X

CeOx and nano-diamond MRF used

additional finishing at LLNL using continuous pitch polishing techniques and CeOx slurry with which 25 μm of fused silica was removed from each of the optical surfaces. Selected optics from these latter types were subjected to MRF final polishing at QED where a uniform layer of fused silica was removed from each of the optic surfaces using CeOx or nano-diamond MR fluid media.

In all experiments, the optics were cleaned using our best-known cleaning procedures²³ prior to etching and optimized 3ω laser conditioning/damage testing. This cleaning procedure included a standard buffing with colloidal alumina as one of the cleaning steps. The etched specimens were prepared at LLNL by immersing the cleaned optics in an aqueous solution of 20:1 low-particulate-grade buffered-oxide etch composed of 38 % (w/w) ammonium fluoride and 2.45 % (w/w) HF at 23 C. After a 57 minutes immersion time, 2 μm of fused silica was removed from both of the optical surfaces of the Zygo-supplied samples. The other optic types were immersed for 28 minutes to remove 1 μm of material from the optical surfaces. The optics were subsequently washed thoroughly using distilled de-ionized water, dried, and cleaned by LLNL personnel using the fused silica optics cleaning procedures described above.

Large area damage testing²⁴ was conducted using a frequency-tripled Spectra-Physics Nd:YAG laser operating at 355-nm with a 7.5-nsec temporal system pulse width and a spot size of ~1mm. Peak laser fluence was determined using the peak fluence measured for a Gaussian-shaped beam spot scaled from the system temporal pulse width to an equivalent 3-nsec wide pulse by $\tau^{1/2}$ scaling, where τ represents the ratio of the pulse lengths. Area scans were conducted with raster step sizes corresponding to the measured Gaussian width at 50% of the peak intensity value. The peak probe fluence was held within +/- 5 % of target via computer control of the laser system’s energy attenuator. Damage detection and quantification was accomplished by side lighting the optic with white light and imaging the area of optic being damage tested onto a scanning linear CCD mega-pixel array²⁴. This diagnostic enabled a digital micrograph to be generated of the area under test that highlighted defects within the bulk and on the optic surface with a resolution of 80-μm/pixel and 10-μm sensitivity. The digital micrograph was subsequently analyzed and defects identified via threshold image processing and counting techniques. The area scaling approach described by Schwartz, *et al.*²⁴, was not used due to the difficulty in selecting Weibull coefficients when no damage can be observed in the tested area. All values are therefore reported as raw, uncorrected damage densities.

For the optics that were not subjected to laser conditioning, large area damage testing was conducted by raster scanning over the test areas while holding the laser fluence constant. For the laser conditioned optics, the specimens were subjected to 3 large area laser scans at a pulse width of 7.5 nsec and peak fluences of 4, 6, and 8 J/cm², each before being probed at laser fluences starting at 10 J/cm². As in the unconditioned experiments, these large area laser raster scans were conducted over the test areas while holding the laser fluence constant.

3. RESULTS AND DISCUSSION

3.1 Zygo ZrOx Conventionally Finished and MRF Finished Fused Silica Optics

The finished optics supplied by Zygo demonstrate the feasibility of using MRF in fabricating 3 ω damage-resistant optics. These experiments provide a direct comparison between the damage densities of the two optics that were prepared identically by lap polishing using ZrOx slurry, one of which was subjected to further finishing by MRF and one of which was not. The experiments also provide information central to the development of the combined MRF, wet etching, and 3 ω post processing procedure used to yield superior performance optics. The results of large area damage testing conducted on each of the specimens are listed in Tables 2 and 3 and plotted in Figures 2 and 3.

Table 2: Damage density observed on 15.2-cm-diameter by 1.0-cm-thick fused silica samples polished by Zygo using ZrOx conventional polishing. Reported damage density is in sites/cm² after illumination by 1 shot/site at 355 nm with no correction for beam shape. Fluences are scaled to equivalent values at 3-nsec.

Fluence (J/cm ²)	Zygo ZrOx As Is	Zygo Conditioned ZrOx	Zygo Etched ZrOx	Zygo Etched - Conditioned ZrOx
4	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00
8	0.00	0.00	0.00	0.00
10	0.32	0.03	0.00	0.00
12	1.00	0.25	0.81	0.02
14	Not Tested	1.00	1.25	0.11

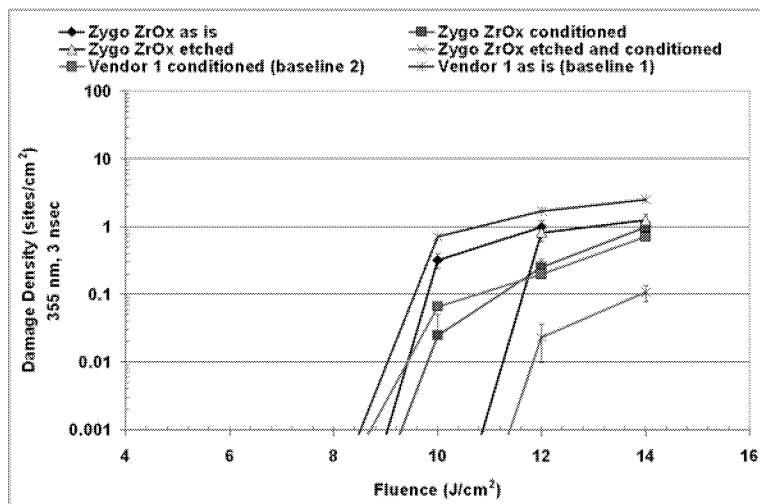


Figure 2: Damage density observed at 355 nm, 3 nsec, on 15.2 cm diameter by 1.0 cm thick fused silica samples polished by Zygo using ZrOx conventional polishing.

Large area damage testing on the Zygo ZrOx conventionally finished parts yields damage densities comparable to those observed on optics that are conventionally finished using the best known methods for 3 ω fused silica fabrication, Table 2 and Figure 2. The comparisons are based upon damage densities taken from measurements made in March 2001 on a 43-cm X 43-cm X 2.5-cm large aperture flat optic supplied by Vendor 1 that represented the best 3 ω optic fabrication procedure known at that time. The unetched and unconditioned Zygo ZrOx finished part (Zygo ZrOx as is) exhibits damage performance about 1.5-2 times better than the unetched and unconditioned Vendor 1 supplied optic (baseline 1). This supports earlier experience that the using ZrOx slurry during the entire conventional finishing process reduces the deleterious effects arising from light absorbing species, such as CeOx slurry, which can be introduced into the

surface polishing and subsurface damage layers. These layers typically contain an admixture of polishing slurry and other contaminants that cause damage when irradiated at high laser fluence. With 3 ω laser conditioning (Zygo ZrOx conditioned), a factor of 3-10 improvement in performance is observed over that of the unetched ZrOx polished optic.

This places its damage performance between typical and the best damage performance observed on large aperture flat optics (Vendor 1 conditioned baseline 2). The improvement is most pronounced at lower laser fluences. This is similar to the factor of 3-8 improvements in damage density that results from conventional CeOx polished optics that are etched. Etching 2 μm from the surfaces of the ZrOx finished optic (Zygo ZrOx etched) improves damage performance to a level equivalent to that observed on parts that undergo 3ω laser conditioning. Laser conditioning at 3ω of the etched ZrOx part (Zygo ZrOx etched and conditioned) yields a performance improvement beyond the best damage performance observed for conditioned high quality optics (Vendor 1 conditioned, baseline 2) by about a factor of 10. The improvements achieved by the combination of etching and 3ω laser conditioning processes on the ZrOx optics suggest that particulate or thin film contamination is present in the polishing and subsurface damage layers which impacts damage performance. Etching is successful in removing or minimizing this contamination and its effects on laser damage performance and 3ω conditioning provides for further improvement by removing what the etching process leaves behind.

Large area damage testing on the Zygo MRF finished parts, Table 3 and Figure 3, yields mixed results ranging from among the worst to the best-performing optic depending upon the post-MRF polishing procedures used.

Table 3: Damage density observed on 15.2-cm-diameter by 1.0-cm-thick fused silica samples polished by Zygo using ZrOx conventional polishing techniques and final polished using MRF to remove 1 μm of material. Reported damage density is in sites/cm² after illumination by 1 shot/site at 355 nm with no correction for beam shape. Fluences are scaled to equivalent values at 3-nsec.

Fluence (J/cm ²)	Zygo 1 μm CeOx MRF As Is	Zygo Conditioned 1 μm CeOx MRF	Zygo Etched 1 μm CeOx MRF	Zygo Etched - Conditioned 1 μm CeOx MRF
4	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00
8	0.00	0.83	0.00	0.00
10	0.56	6.70	0.00	0.00
12	11.40	Gray Haze	0.06	0.01
14	Gray haze	Not Tested	1.06	0.02

Significant damage densities are observed on the unetched Zygo CeOx MRF finished optics when tested under both unconditioned and 3ω laser conditioned procedures. The unetched and unconditioned MRF optic (Zygo 1 μm CeOx MRF as is) possesses unacceptable damage density of 0.56 sites/cm² at 10 J/cm² and 11.40 sites/cm² at 12 J/cm². Damage is even more severe during and after 3ω laser conditioning with the conditioned MRF part (Zygo conditioned 1 μm CeOx MRF) displaying damage densities near 0.83 sites/cm² at 8 J/cm² and 6.70 sites/cm² at 10 J/cm². 3ω laser conditioning in this instance is counterproductive. Such results are unexpected based upon previous experience with improvements gained by laser conditioning on conventionally finished optics. As such, damage testing was not conducted at higher fluences due to the risk of catastrophic failure. The poor performance of the unetched MRF optics can most likely be attributed to optically absorbing iron and CeOx media used by the MRF tool during finishing. These materials have a high probability of combining with, or being present in, the optic surfaces as thin films or particulate inclusions in the surface polishing and subsurface damage layers. Etching 2 μm from the surfaces of the MRF polished specimen yields a significant improvement in damage performance. Testing of the etched MRF finished optics (Zygo etched 1 μm CeOx MRF) results in damage performance equivalent to that observed for the

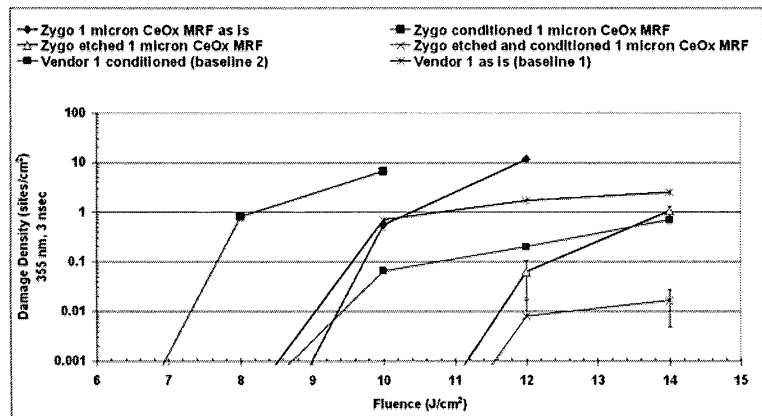


Figure 3: Damage density observed at 355 nm, 3 nsec, on 15.2 cm diameter by 1.0 cm thick fused silica samples polished by Zygo using ZrOx conventional polishing techniques and final polished using MRF to remove 1 μm of material.

etched and unconditioned ZrOx finished parts, unetched and 3 ω laser conditioned ZrOx finished parts, and conditioned high quality optics (Vendor 1 conditioned, baseline 2). Again, the improvement brought about by etching is more pronounced at lower laser fluences. Combining the etching process with 3 ω laser conditioning (Zygo etched and conditioned 1 μm CeOx MRF) yields damage performance superior to any previously observed on high quality conventionally finished large aperture optics (Vendor 1 baselines 1 and 2). The damage density observed using this combined procedure is 0.02 sites/cm² at 14 J/cm² which is about a factor of 50-150 times lower than the densities observed on Vendor 1 supplied optics. More important, damage is not observed at fluences less than 12 J/cm², and only a single damage site observed at 12 J/cm² over a 122-cm² analysis area. This behavior has not been previously observed on any conventionally finished optic tested at LLNL. The damage density at 14 J/cm² is also impressive with only 2 cumulative damage sites observed over the same analysis area. The damage test results suggest that etching the surface of the optics removes MRF-induced contamination (iron and/or CeOx) present in a thin polishing layer. The removal of this layer by etching and 3 ω laser conditioning leaves a new exposed surface free from contamination and few to no pits and cracks that could interact with the laser probe to induce damage.

Chemical analysis of the etch solutions used to process the Zygo ZrOx conventionally polished and MRF optics gives an indication of the polishing layer composition that is removed from the optical surfaces by etching. For the ZrOx polished parts, 8.15 ppmw of zirconium, 0.46 ppmw of cerium, and 1.62 ppmw of iron are measured with respect to the fused silica removed in the etch solutions after removal of 2 μm of SiO₂ from the optical surfaces via etching. These assays provide evidence indicating that conventional polishing leaves a polishing layer and potentially a subsurface damage layer at or near the optical surfaces which is composed of an admixture of materials contained in the polishing slurry and from other contaminants from other polishing and finishing steps. Removal of these contaminants using wet acid etching explains the improvement in damage performance observed as both of the contaminants absorb strongly in the ultraviolet. For the MRF-finished optics, 5.67 ppmw of cerium and 5.36 ppmw of iron are measured with respect to the fused silica removed in the solutions after removal of 2 μm of SiO₂ from the optical surfaces via etching. The final MRF polish appears to successfully remove the zirconium in the optical surfaces left behind by conventional polishing but generates a new polishing layer containing MR fluid components. The amounts of iron and cerium measured in this instance shows that contamination by absorbing species at ppmw levels are sufficient to deteriorate the optic's damage

resistance to high laser fluences. This explains the poor performance observed on the unetched MRF finished parts regardless of conditioning and partially explains the improvements observed after etching is performed.

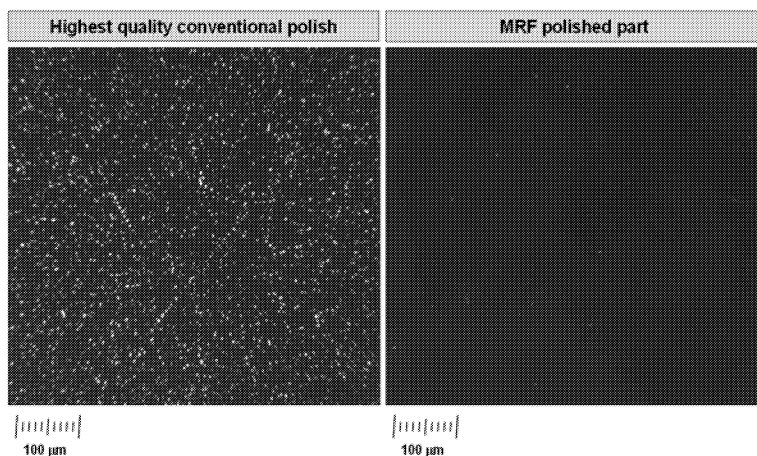


Figure 4: After etching, comparison of dark-field microscopy on highest quality, conventionally polished parts and MRF polished parts illustrate the dramatic decrease in scatter sites and “chatter marks” normally associated with subsurface damage from conventional finishing.

By its very nature, MRF possesses a unique capability for fabrication of optics with low subsurface damage. This can be attributed to small normal stress loading on a glass surface compared to shear stress which is created by the interaction of the tool's magnetic field and the MR fluid through the converging gap between the optic and tool's pole faces. Thus, the shear stress applied to the optic results in high material removal through the use of CeOx abrasives while at the same time avoiding medial crack formation and propagation. The damage test data suggests that this is the case and that only slight surface mixing occurs where the iron and CeOx exist

on the optic surfaces and can be easily removed with etching. Dark-field microscopy of the etched parts shows that MRF finishing results in a dramatic reduction in subsurface damage when compared to that present on conventionally polished parts, Figure 4. The large number of scatter sites and “chatter” on the conventionally lapped optical surfaces suggests that the subsurface damage layer is significant in spite of care taken to manage the damage during polishing. It also suggests that the large number of sites lead to a high probability for retention of absorbing species that can initiate

damage when irradiated at high fluence. In contrast, the etched MRF polished optical surfaces show a near absence of subsurface damage that explains the large improvement observed in damage performance when compared to the best performing conventionally polished optics.

3.2 MRF Final Finishing and 3ω Post Processing of Vendor 1 Conventionally Finished Fused Silica Optics

The experiments involving the application of various combinations of MRF final polishing, wet etching, and 3ω post processing techniques to subsections cut from the fused silica flat provided by Vendor 1 are intended to show that improvements in damage performance can be made beyond that observed for the best performing large aperture conventional finishing process. As in the case for the Zygo-prepared optics, these experiments allow for direct comparison of the combined 3ω finishing process on optics that are fabricated using the same process steps. The experiments are also meant to test if different MR fluid media and MRF removal amounts can impact the optical damage performance. In these instances, MR fluids containing nano-diamond and CeOx polishing media are examined. For MRF final finishing using CeOx MR fluid, two different removal amounts are explored which include 0.25 and 1 μm material removal from both optical surfaces.

The results of large area damage testing conducted on each of the specimens are listed in Table 3 and plotted in Figure 5. Etching 1 μm from the surfaces of the Vendor 1 supplied optic results in about a factor of 4 improvement in the damage

Table 3: Damage density observed on 14.3-cm X 14.3-cm X 2.5-cm fused silica optic cut from full size NIF flat optic polished by Vendor 1. Reported damage density is in sites/cm² after illumination by 1 shot/site at 355 nm with no correction for beam shape. Fluences are scaled to equivalent values at 3-nsec.

Fluence (J/cm ²)	Vendor 1 Etched	Vendor 1 Etched - Conditioned	Vendor 1 Conditioned 1 μm nano-diamond MRF	Vendor 1 Etched - Conditioned 1 μm nano-diamond MRF	Vendor 1 Etched - Conditioned 0.25 μm CeOx MRF	Vendor 1 Etched - Conditioned 1 μm CeOx MRF
4	0.00	0.00	0.00	0.00	0.00	0.00
6	0.00	0.00	0.00	0.00	0.00	0.00
8	0.00	0.02	4.04	0.00	0.00	0.00
10	0.17	0.04	Gray Haze	0.02	0.01	0.01
12	0.43	0.07	Not Tested	0.07	0.06	0.02
14	0.65	0.23	Not Tested	0.14	0.23	0.08

performance over that observed for the unprocessed part (Vendor 1 as is baseline 1). Applying 3ω laser conditioning to the etched optic further improves the part performance by about a factor of 11, with the optic performing about a factor of 3 better than the Vendor 1 conditioned optic, baseline 2. The increase in damage resistance produced by etching and 3ω laser conditioning is consistent with the performance enhancement observed on the Zygo supplied parts and shows that 3ω post processing itself can be a valuable tool to minimize damage initiation under high fluence conditions.

Final polishing of the Vendor 1 optics using MRF with nano-diamond MR fluid to remove 1 μm of SiO₂ from the optical surfaces yields results similar to that observed for the Zygo-supplied specimens. Again, large area damage testing on the optics yields mixed results ranging from unacceptable for high fluence laser application to superior performance depending upon the post-MRF procedures used. For the unetched and laser conditioned nano-diamond polished MRF optic, significant damage densities are observed at low fluence, 4.04 sites/cm² at 8 J/cm² with the appearance of gray haze at 10 J/cm². Additional testing of the unetched optic and unconditioned optic was not pursued as it is expected to perform poorly based upon the results of the Zygo provided samples. Etching 1 μm of SiO₂ from the surfaces of the nano-diamond MRF polished samples and application of 3ω laser conditioning leads to an improvement in damage performance equivalent to simply etching and laser conditioning the Vendor 1 supplied optic. Larger damage sites are observed, however, for these optics when compared to the sites observed on conventionally polished and CeOx MRF polished parts. Even though the nano-diamond portion of the MR fluid is not expected to interact with the polishing layer on the optical surfaces as is the case for CeOx MR fluid media, the interaction occurring at the glass surface apparently leaves an admixture of iron and diamond in the polishing layer. This explains the unacceptable performance that is observed on the conditioned part and the damage resistance improvements realized after etching. Etching appears to be effective at removing most of the contamination present but seems to leave behind nano-diamond remnants that promote larger damage sites when interacting with the laser beam at high fluence. As the nano-diamonds used in the MR

fluid are anthropogenic, it is possible that they contain contaminants that can contribute to damage observed in the fused silica parts even though one would conclude that the diamond itself does not contribute. Furthermore, it is possible that contaminants in the nano-diamonds are initiating damage to the diamonds that subsequently results in collateral damage to the glass.

MRF removal of 0.25 μm of fused silica from the surfaces of the Vendor 1 optics using CeOx MR fluid followed by etching of 1 μm from the optical surfaces and 3ω laser conditioning results in damage performance improvement equivalent to that observed on the 1 μm nano-diamond MRF and 3ω post-processed specimens. In this case, the damage performance is about a factor of 3-4 times better than the best performing conventionally finished optic after undergoing

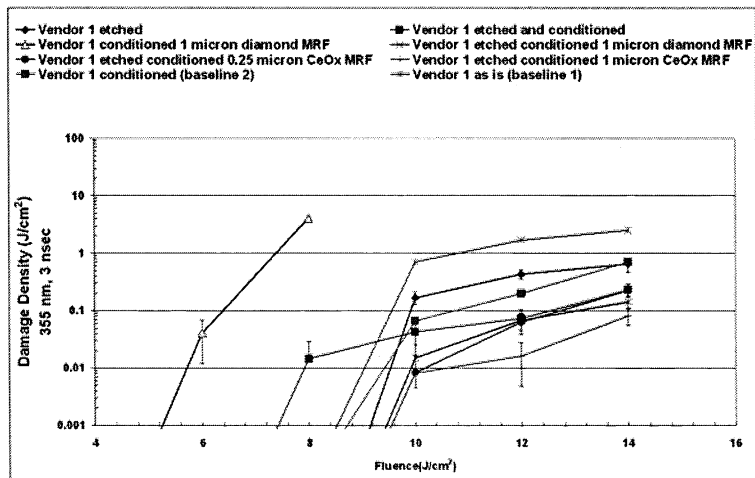


Figure 5: Damage density observed at 355 nm, 3 nsec, on 14.3-cm x 14.3-cm x 2.5-cm fused silica optic cut from full size NIF flat optic polished by Vendor 1.

3ω laser conditioning, Vendor 1 Conditioned, Figure 4. MRF removal of 1 μm of fused silica from the optical surfaces using CeOx MR fluid and 3ω post-processing provides nearly a factor of 9-12 improvement. The improvement, however, is not as large as that observed on the Zygo provided MRF polished specimens, 0.08 sites/cm² versus 0.02 sites/cm². This observation could lead to the conclusion that the quality of the optic before MRF has some effect on its ultimate laser damage performance. It is highly likely that the ultimate damage performance of the optic depends upon the subsurface damage layer thickness and the contaminants residing therein and that the MRF/ 3ω post-processing procedure may need to be optimized to ensure that sufficient material removal is accomplished to realize ultimate and reliable performance. Further experiments in this area are necessary in order to confirm the observation and form a sound conclusion.

3.3 MRF Final Finishing and 3ω Post Processing of Vendor 2 Conventionally Finished Fused Silica Optics

Application of MRF final polishing and 3ω post processing techniques to subsections cut from the fused silica flat provided by Vendor 2 are meant to illustrate that improvements in damage performance can be made on optics that are unacceptable for high fluence ultraviolet laser applications. These represent optics for which little to no attention was paid to the type of polishing slurry used nor to the extent of subsurface damage encountered during the fabrication process. The results of large aperture damage testing on the subsections are presented in Table 4 and Figure 6.

The damage performance observed on the unetched and unconditioned part (Vendor 2 as is) identifies its usefulness as marginal with a damage density of about 0.2 sites/cm² at fluences in the range of 4-8 J/cm². At laser fluences above 10 J/cm², the damage density drastically increases and quickly becomes severe with a value of 314 sites/cm² at 14 J/cm². Damage densities at these levels make the part unacceptable for use due to the risk of catastrophic failure from damage growth. The damage morphology is recognizable and typical for a part that has been conventionally finished using CeOx lap techniques. The optical surface appears “hazy” with a large number of pinpoint fractures indicating that the surface contains contaminated polishing and subsurface damage layers that are laden with contaminants. Etching 1 μm from the surfaces or conducting laser conditioning improves the damage performance by about a factor of 100, however, either treatment only improves the optic to a point equivalent to that observed on the Vendor 1 unetched and unconditioned part (Vendor 1 as is, baseline 1). Combined etching and 3ω laser conditioning improves the conventionally finished part by a factor of 300 with values in the vicinity of 0.2-1 sites/cm² between 10 and 14 J/cm².

Table 4: Damage density observed on 14.3-cm X 14.3-cm X 1.0-cm fused silica optic cut from full size NIF high aspect ratio optic polished by Vendor 2. Reported damage density is in sites/cm² after illumination by 1 shot/site at 355 nm with no correction for beam shape. Fluences are scaled to equivalent values at 3-nsec.

Fluence (J/cm ²)	Vendor 2 As is	Vendor 2 Conditioned	Vendor 2 Etched	Vendor 2 Etched and Conditioned	Vendor 2 Etched - Conditioned 1 μm CeOx MRF	Vendor 2 Etched - Conditioned 1 μm nano-diamond MRF
4	0.13	0.00	0.01	0.00	0.00	0.00
6	0.24	0.00	0.01	0.00	0.00	0.00
8	0.29	0.38	0.31	0.00	0.00	0.02
10	0.46	0.48	0.47	0.18	0.02	0.04
12	10.00	1.34	1.43	0.50	0.02	0.27
14	314.00	2.66	2.86	1.05	0.18	0.52

This damage density is still about a factor of 2-3 larger than that observed on the best performing conventionally finished large aperture optic with and 3ω laser conditioning (Vendor 1 conditioned, baseline 2). Etching and laser conditioning appear to reduce the damage density to a point depending upon the initial quality of the optic, which is in this case is poor, but reach a floor where they can not bring about any more improvement. This is consistent with the observations noted on the other optic families tested.

Application of MRF final polishing to the Vendor 2 conventionally finished optics using either nano-diamond or CeOx MR fluid to remove 1 μm of fused silica from the polished surfaces show improvements in damage resistance when combined with the 3ω post processing procedure. After etching of 1 μm from the optic surfaces and application of 3ω laser conditioning, the Vendor 2 nano-diamond MRF polished optic shows about a factor of 600 improvement compared to the untreated part. The damage density is equivalent to the performance observed for the best performing conventionally finished large aperture optic with and 3ω laser conditioning (Vendor 1 conditioned, baseline 2). A factor of 1700 improvement is obtained using CeOx MRF polishing and the full advanced finishing treatment which yields superior performance 4 times better than that measured for the Vendor 2 conditioned optic. This trend is consistent to that noted in the Vendor 1 MRF finishing experiments. CeOx MR fluid appears to be more effective than nano-diamond MR fluid in removing the subsurface damage layer without leaving behind remnants which promote damage. A major conclusion drawn from the experiments on the Vendor 2 supplied optics is that significant improvements can be made on poorly performing conventionally finished optics by using etching and 3ω laser conditioning; however, when combined with MRF final polishing, the optics can be made into superior performers. Like in the Vendor 1 experiments, the experiments on the Vendor 2 optics indicate that the quality of the optic prior to MRF final polishing may be important in determining the ultimate damage performance due to the thickness of the subsurface damage layer resulting from the conventional finishing process. The MRF removal may have to be optimized for the specific processes used for finishing before MRF final polishing to ensure that a sufficient amount of material is removed from the surfaces to get past the subsurface damage.

Application of MRF final polishing to the Vendor 2 conventionally finished optics using either nano-diamond or CeOx MR fluid to remove 1 μm of fused silica from the polished surfaces show improvements in damage resistance when combined with the 3ω post processing procedure. After etching of 1 μm from the optic surfaces and application of 3ω laser conditioning, the Vendor 2 nano-diamond MRF polished optic shows about a factor of 600 improvement compared to the untreated part. The damage density is equivalent to the performance observed for the best performing conventionally finished large aperture optic with and 3ω laser conditioning (Vendor 1 conditioned, baseline 2). A factor of 1700 improvement is obtained using CeOx MRF polishing and the full advanced finishing treatment which yields superior performance 4 times better than that measured for the Vendor 2 conditioned optic. This trend is consistent to that noted in the Vendor 1 MRF finishing experiments. CeOx MR fluid appears to be more effective than nano-diamond MR fluid in removing the subsurface damage layer without leaving behind remnants which promote damage. A major conclusion drawn from the experiments on the Vendor 2 supplied optics is that significant improvements can be made on poorly performing conventionally finished optics by using etching and 3ω laser conditioning; however, when combined with MRF final polishing, the optics can be made into superior performers. Like in the Vendor 1 experiments, the experiments on the Vendor 2 optics indicate that the quality of the optic prior to MRF final polishing may be important in determining the ultimate damage performance due to the thickness of the subsurface damage layer resulting from the conventional finishing process. The MRF removal may have to be optimized for the specific processes used for finishing before MRF final polishing to ensure that a sufficient amount of material is removed from the surfaces to get past the subsurface damage.

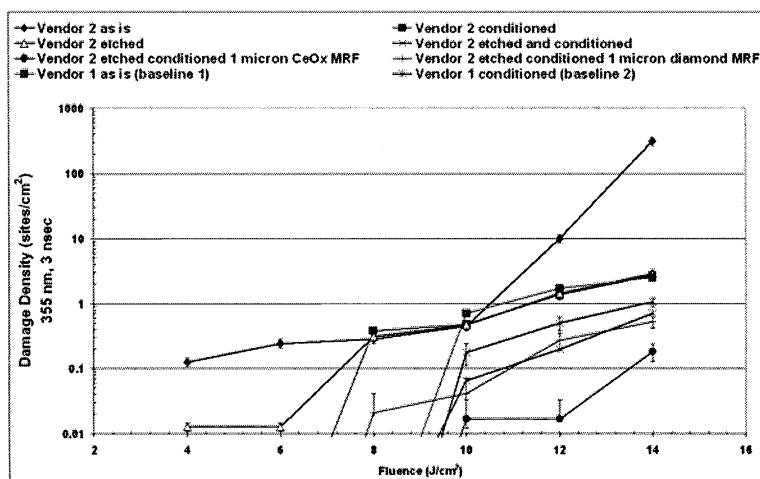


Figure 6: Damage density observed at 355 nm, 3 nsec, on 14.3-cm x 14.3-cm x 1.0-cm fused silica optic cut from full size NIF high aspect ratio optic polished by Vendor 2.

3.4 MRF Final Finishing and 3ω Post Processing of LLNL Conventionally Finished Fused Silica Optics

Additional CeOx lap polishing of subsections cut from the Vendor 2 supplied conventionally finished optics is intended to test the possibility of improving performance through careful removal of fused silica from the optical surfaces prior to MRF final polishing. The goal in these experiments is to remove material under small normal loads to remove or minimize the subsurface damage layer present from prior fabrication steps. A further assumption is that additional lapping of 25 μm from the optical surfaces is sufficient to eliminate these effects. The results of large aperture damage testing on the subsections are presented in Table 5 and Figure 7.

Table 5: Damage density observed on 14.3-cm X 14.3-cm X 1.0-cm fused silica optic cut from full size NIF high aspect ratio optic polished by Vendor 2 and subsequently polished at LLNL using subsurface damage management. Reported damage density is in sites/cm² after illumination by 1 shot/site at 355 nm with no correction for beam shape. Fluences are scaled to equivalent values at 3-nsec.

Fluence (J/cm ²)	LLNL CeOx Etched and Conditioned	LLNL CeOx Etched - Conditioned 1 μm CeOx MRF	LLNL CeOx Etched - Conditioned 1μm nano-diamond MRF
4	0.00	0.00	0.00
6	0.00	0.03	0.04
8	0.00	0.03	0.06
10	0.05	0.05	0.16
12	0.23	0.11	0.29
14	0.53	0.20	0.53

Overall, the results of etching, 3ω laser conditioning, and combined MRF/3ω post processing are similar to those obtained for the Vendor 2 supplied optics. Etching 1 μm from the surfaces of the optic and applying 3ω laser conditioning yields damage performance equivalent to that observed on the best performing conventionally finished

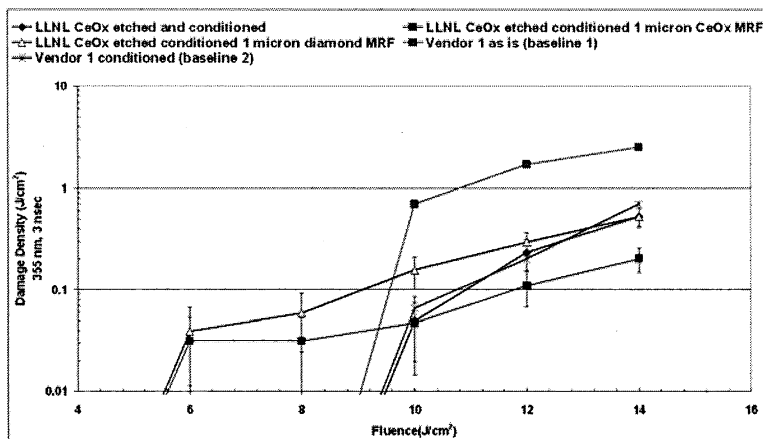


Figure 7: Damage density observed after illumination by 1 shot/site at 355 nm with no correction for beam shape. Fluences are scaled to equivalent values at 3-nsec.

large aperture optic with 3ω laser conditioning (Vendor 1 conditioned, baseline 2). This performance is a factor of 2 better than the Vendor 1 etched and conditioned optic which suggests that the additional material removal from the lapping process was successful at reducing the subsurface damage layer thickness. Application of MRF final polishing to the LLNL CeOx conventionally finished optics using either nano-diamond or CeOx MR fluid to remove 1 μm of fused silica from the polished surfaces show improvement in damage resistance when combined with the 3ω post processing procedure. A comparison of nano-diamond and CeOx MR fluid shows that the CeOx based fluid results in optics with superior UV damage resistance. For the CeOx MRF polished sample, the damage performance is about 3-4 times better than that observed on the Vendor 1 conditioned part. This is consistent with the trend observed on

the other optic families tested from Vendor1 and Vendor 2. On the whole, these experiments suggest that additional lap polishing creates new damage or chases existing damage in the optics and that the combined MRF/3ω post processing procedure is more effective and efficient at removing the deleterious effects of prior optical fabrication processes to bring about superior damage performance.

4. CONCLUSIONS

The large area damage tests conducted on optics finished by a number of different vendors show that MRF, when combined with appropriate 3ω post-processing, significantly reduces the damage sensitivity for high fluence UV laser light. This is true regardless of the quality of the original conventional finishing. MRF can successfully remove subsurface damage and the contaminants contained therein leaving a high quality surface with little to no subsurface pits and cracks that could interact with the laser beam to induce damage. Even though MRF is successful at removing subsurface damage, the final MRF polish generates a new polishing layer containing MR fluid components at levels that are sufficient to promote damage. The MRF-induced polishing layer can be successfully removed via etching and laser conditioning to yield superior performance. This is in contrast to the subsurface layer generated by conventional polishing which contains debris and a large number of pits and cracks which lead to a high probability for retention of optically absorbing species that can initiate damage when irradiated at high fluence. The MRF finishing process shows promise when combined with etching and 3ω laser conditioning in decreasing the damage concentration to levels that can be managed by mitigation procedures currently being considered for use on high laser damage threshold optical flats as well as continuous phase plates. Application of either nano-diamond or CeOx MRF final polishing results in significant damage performance improvement, with the CeOx MRF process being superior. Additionally, nano-diamond MRF polished parts possess more numerous damage sites that are larger in size. The present experiments also show that the quality of the optic before MRF has some effect on the ultimate performance of the optic after application of the combined MRF/ 3ω post-processing procedure. Ultimate and reliable performance depends on the amount of subsurface damage present which suggests that the MRF process needs to be optimized to the specific processes used for optical fabrication to ensure that a sufficient amount of material is removed. The combined MRF/ 3ω post-processing procedure is flexible. It can be used during initial optic fabrication to provide for superior damage performance or it can be used on existing lesser quality conventionally finished parts to improve performance beyond that attainable by using conventional techniques and post processing alone.

5. ACKNOWLEDGEMENTS

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REFERENCES

1. A.K. Burnham, L. Hackel, P. Wegner, T. Parham, L. Hrubesh, B. Penetrante, P. Whitman, S. Demos, J. Menapace, M. Runkel, M. Fluss, M. Feit, M. Key, T. Biesiada, "Improving 351-nm damage performance of large-aperture fused silica and DKDP optics", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
2. A. M. Rubenchik and M. D. Feit, "Initiation, growth, and mitigation of UV damage in fused silica", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
3. A. Salleo, R. Chinsio, F. Y. Genin, "Crack propagation in fused silica during UV and IR ns-laser illuminations", *Laser Induced Damage in Optical Materials, SPIE Proc.* **3578**, 456-471 (1998).
4. J. Murray, "Overview of the National Ignition Facility", *SPIE* **3492** supplement, 1-10 (1998).
5. B. M. Van Wonterghem, J. R. Murray, J. H. Campbell, S.R. Speck, C. E. Barker, I. C. Smith, B.F. Browning, W.C. Behrendt, "Performance of a prototype for a large-aperture multi-pass Nd:glass laser for inertial confinement fusion", *Applied Optics*, **36**, 4932-4952 (1997).
6. R. M. Brusasco, B. M. Penetrante, J. A. Butler, and L. W. Hrubesh, "Localized CO₂ laser treatment for mitigation of 3ω damage growth in fused silica", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
7. B. M. Penetrante, R. M. Brusasco, J. A. Butler, S. M. Maricle, and J. E. Peterson, "CO₂ laser polishing for reduction of 3ω damage initiation in fused silica", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
8. L. W. Hrubesh, M. A. Norton, W. A. Molander, E. E. Donohue, S. M. Maricle, B. M. Penetrante, R. M. Brusasco, W. Grundler, J. A. Butler, J. W. Carr, R. M. Hill, L. J. Summers, M. D. Feit, A. Rubenchik, M. H. Key, P. J. Wegner, A. K. Burnham, L. A. Hackel, and M. R. Kozlowski, "Methods for mitigating surface damage growth in NIF final optics", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).

9. B. M. Penetrante, J. E. Peterson, S. M. Maricle, and R. M. Brusasco, "UV laser conditioning for reduction of 3 ω damage initiation in fused silica", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
10. Z. Gu, P. Liang, and W. Zhang, "Influence of glass surface layers on laser-induced damage threshold", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
11. P. Bouchut, P. Garrec, J. Newport, P. Gacoin, F. Bonnemason, S. Kaladgew, "Technological processing of silica as seen through laser induced damage tests", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
12. M. J. Fluss, J. Plitzko, M. Wall, "Initiation identification in 355-nm optics", *Laser Induced Damage in Optical Materials*, this proceedings, Boulder (2001).
13. T. Hoshino, Y. Kurata, Y. Kurata, Y. Terasaki, and K. Susa, "Mechanism of polishing of SiO₂ films by CeO₂ particles", *Journal of Non-Crystalline Solids*, **283**, 129-136 (2001).
14. D. W. Camp, M. R. Kozlowski, L. M. Sheehan, M. Nichols, M. Dovik, R. Raether, I. Thomas, "Subsurface damage and polishing compound affect at the 355-nm laser damage threshold of fused silica surfaces", *Laser Induced Damage in Optical Materials, SPIE Proc.* **3244**, 356-364 (1997).
15. D. Golini, S. Jacobs, W. Kordonski, and P. Dumas, "Precision optics fabrication using magnetorheological finishing", *Advanced Materials for Optics and Precision Structures*, **CR67**, 251-274 (1997).
16. A. B. Shorey, S. D. Jacobs, W. I. Kordonski, and R. F. Gans, "Experiments and observations regarding mechanisms of glass removal in magnetorheological finishing", *Applied Optics*, **40**, No. 1, 20-33 (2001).
17. W. Kordonski, D. Golini, P. Dumas, and S. Jacobs, "Magnetorheological suspension-based finishing technology", *SPIE*, **3326**, 527-535 (1998).
18. S. D. Jacobs, D. Golini, Y. Hsu, B. E. Puchebner, D. Stafford, W. I. Kordonski, I. V. Prokhorov, E. Fess, D. Pietrowski, and V. W. Kordonski, "Magnetorheological finishing: a deterministic process for optics manufacturing", *SPIE*, **2576**, 372-382 (1995).
19. S. D. Jacobs, F. Yang, E. M. Fess, J. B. Feingold, B. E. Gillman, W. I. Kordonski, H. Edwards, and D. Golini, "Magnetorheological finishing of IR materials", *SPIE*, **3134**, 258-269 (1997).
20. S. R. Arrasmith, I. A. Kozhinova, L. L. Greg, A. B. Shorey, H. J. Romanofsky, S. D. Jacobs, D. Golini, W. I. Kordonski, S. Hogan, and P. Dumas, "Details of the polishing spot in magnetorheological finishing", *SPIE*, **3782**, 92-100 (1999).
21. D. Golini, W. I. Kordonski, P. Dumas, and S. Hogan, "Magnetorheological finishing (MRF) in commercial precision optics manufacturing", *SPIE*, **3782**, 80-91 (1999).
22. G. A. C. M. Spierings, "Review: Wet chemical etching of silicate glasses in hydrofluoric acid based solutions", *Journal of Materials Science*, **28**, 6261-6273 (1993).
23. S. Frieders, "Precision cleaning of bare fused silica optics", LLNL Draft Processing Procedure, (2001).
24. S. Schwartz, M. D. Feit, M. R. Kozlowski, and R. P. Mouser, "Current 3 ω large optic test procedures and data analysis for quality assurance of National Ignition Facility optics", *Laser Induced Damage in Optical Materials, SPIE Proc.*, **3578**, 314-321 (1998).

Combined advanced finishing and UV-laser conditioning for producing UV-damage-resistant fused silica optics (4679-65)

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Q. When you etch, one or a couple microns, do you have any problems with increasing the roughness of the surface?

A. We've measured the zygo parts. We've started with the micro roughness roughly around 3 1/2 angstroms, and after etching two microns of material away from the surface, we are up around 3.8 and 3.9 angstroms so the quality of the optics is maintained even though we are etching two microns away from the surface. A lot of this has to do with the care that's taken in the preparation of the optic through grinding and polishing to help mitigate the cracks and the surface finish that is on the bad optics. If you start with a bad optic, once you get rid of the polishing layer you will dramatically increase the roughness and the short frequency waviness. We also have very good short frequency waviness on these optics; roughly about .7 nm rms, about a factor of three below our specs.

Q. First of all I would like to congratulate you on great progress. Have you studied the effect of processing on growth behavior? Apparently it is more difficult to initiate damage. How about the growth?

A. The work that I'm talking about here basically pays attention to the initiation sites. There's other work if you go look at the posters, for example, the poster paper of Bernie Penetrony. We've seen some improvement, but not total improvement with HF etching. If you get rid of everything with HF etching once you have damage site. If you etch it, sometimes they grow and sometimes they don't. We don't fully understand what's going on there. Some improvement is shown with etching. What we've seen with the MRF damage sites using the cerium oxide MR fluid is that the damage sites are smaller than what we've typically seen in the past with conventional polishing. We are getting fewer damage sites in number, but when it does damage, the damage sites are comparatively smaller unless we use diamond, and those happen to be significantly larger.

Q. If you use RMF on a large-scale part – and if you did – how did you measure the flatness of the surface? As I am aware, RMF isn't really very well tuned to the large part, and it also shows the influence of the so-called mid range frequencies to several tenths of the micrometer scale like it is creating such an additional frequency.

A. We are in the process at this point of doing that. Zygo is actually performing finishing using MRF at this time on full size NIF parts. We are hoping in the next month or two to have a first look at the full size optic at NIF with regard to how flat it is, what our gradients are, and how our waviness is and how that compares to what we have conventionally finished up to this point and also paying attention to the damage performance of the optic. The answer is that we don't have any information on a large part, but that is forthcoming.