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Damage Resistant Optical Glasses for High Power Lasers: A Continuing Glass Science and Technology Challenge

J.H. Campbell

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Damage Resistant Optical Glasses for High Power Lasers:

A Continuing Glass Science and Technology Challenge

John H. Campbell
University of California
Lawrence Livermore National Laboratory
P.O. Box 808, L-491
Livermore, California 94550
(campbell12@llnl.gov)

ABSTRACT

A major challenge in the development of optical glasses for high-power lasers is reducing or eliminating laser-induced damage to the interior (bulk) and the polished surface of the glass. Bulk laser damage in glass generally originates from inclusions. With the development of novel glass melting and forming processes it is now possible to make both fused silica and a suit of meta-phosphate laser glasses in large sizes ($\gtrsim$0.5-1m diameter), free of inclusions and with high optical homogeneity ($\sim 10^{-6}$). Considerable attention also has been focused on improving the laser damage resistance to polished optical glass surfaces. Studies have shown that laser-induced damage to surfaces grows exponentially with the number of shots when illuminated with nano-second pulses at 351-nm above a given fluence threshold. A new approach for reducing and eliminating laser-induced surface damage relies on a series of post-polishing treatment steps. This damage improvement method is briefly reviewed.

I. Introduction

To a large extent the development of high-peak-power laser systems has been made possible by corresponding developments of optical glasses. Specifically, improved or new laser glass compositions [1-3], advances in optical glass melting methods [4-8] and novel glass fabrication technologies [9-12] have enabled development of glasses for use in lasers capable of producing megajoules of energy at peta-watt power levels [13,14]. For example, the 192-beam National Ignition Facility currently under construction at Lawrence Livermore National Laboratory (LLNL) is capable of generating energies up to 3.0 MJ at 1053 nm and, after frequency conversion, 1.8 MJ at the third harmonic (351 nm) [13]. At the nominal 3-ns laser operating pulse lengths these energies correspond to peak-power levels of 1.0 and 0.6×10^{15} W, respectively. This laser, like its lower energy and lower peak-power forerunners, is being used for basic research in high-energy-density plasma physics [15] and nuclear fusion energy [16]. In fact, it is the goal of this facility to achieve controlled nuclear fusion ignition and gain in a laboratory setting [17].

Although there are many technological and scientific challenges to developing optical glasses for high-power lasers, perhaps the most critical challenge has been the reduction and, where possible, elimination of laser-induced damage [18-26]. This is a continuing challenge because as new laser-damage-resistant optical materials and fabrication technologies are developed, laser designers increase the system operating energies and powers to the limits of these new materials. The reason, of course, is simply economics; the higher the damage resistance of the optics, the greater the laser output energy that can be achieved for a given investment.

To understand laser-induced damage in optical glasses requires knowledge about many fundamental aspects of these materials; for example: (1) the mechanisms for light interaction with bulk glass and glass surfaces at high intensity and short pulse lengths (fs to ns), (2) the chemistry and physics of glass surfaces as modified by glass finishing ("polishing"), (3) glass fracture and fracture analysis, and (4) radiation-induced defect formation in glass across a broad range of energies. Therefore, major improvements in high power laser system design and performance requires corresponding advances in glass science.

This paper specifically reviews recent advances for producing better (more laser damage resistant) optical glasses for use in high power lasers as well as important areas where further research is needed. The manuscript is divided into three sections. The next section (II) reviews progress on improving the bulk (interior) damage threshold of optical glasses while section III reviews recent theoretical and experimental results toward understanding damage to polished glass surfaces. The final section discusses a novel approach now being used to improve the damage resistance of fused silica for high-power laser applications in the near UV.

Note that laser-induced damage to optical materials has been a subject of intense study for more than 30 years. Therefore it is impossible to fully cover all aspects of laser damage in glasses in this brief review. An annual scientific symposium, beginning in 1971 and extending to today, provides a comprehensive compilation of scientific work in this area [21].

II. Improving the internal damage threshold of optical glasses

Laser damage to the bulk (interior) of a glass is caused by multiphoton-ionization and avalanche breakdown [21]. The fluence necessary to achieve avalanche breakdown is in excess of several hundred Joules/cm^2 for lasers operating at energies well below the material band gap and at nano-second pulse lengths [25]. Therefore the glass surface generally damages first. The exceptions are damage due to self-focusing in the bulk due to self-phase modulation at high intensities and long optical pathlengths and/or damage due to bulk-impurities (e.g. inclusions).

To avoid self-focusing damage, laser designers try to use optical glasses having a low non-linear index, n_2 [18,27-29]. Thus fluoride glasses containing low atomic number modifiers (e.g. BeF_2 [28]) clearly would make the ideal choice due to their intrinsically low index and optical dispersion (and thus low n_2) [29]. Unfortunately most of these glasses are not economically practical and/or can not be melted free of bulk inclusions. Therefore, the development of low-n_2 glasses that can be manufactured free of impurity inclusions would represent a significant advancement in optical glasses for use in high-power laser applications.

Apart from self-focusing, the major cause of bulk damage to glasses is microscopic (□10μm) absorbing inclusions. This problem was recognized early in the development of Q-switched lasers [21,30] and in the 1980's proved to be a major limitation in the further development of both high-peak-power and high-average-power lasers [8,18,26]. The inclusions originate from the melting container materials, particularly platinum. Platinum-lined melting vessels are required to achieve ppm-level optical homogeneity for glasses used in laser applications, however it leads to trace concentrations of microscopic Pt metal particles in the glass [4,8]. The presence of high levels of Pt inclusions in silicate, borosilicate, fluorophosphates, etc. glasses caused laser designers to specify the use of the only two glasses that could be made essentially free of inclusions: (1) a limited number of meta-phosphate glasses for use as active materials (laser gain medium and faraday rotators[1,4]), and (2) CVD-deposited fused silica for passive optical elements (lenses and windows[7,20]). Phosphate laser glasses were chosen because of their inherently high Pt solubilities (~1000 ppm) when melted under oxidizing conditions [8,18,31]. Thus the solution to Pt inclusions is to carefully control the melting conditions so the small numbers of residual Pt particles dissolve into the phosphate glass matrix [4,32-34]. This discovery has enabled the construction of a series of high-peak-power lasers beginning in the mid-80's [35-39].

Fused silica made by conventional CVD processing [7] proved adequate for the operating fluences of the lasers that were constructed and operated in the 80's and 90's. However, the National Ignition Facility (NIF) has much higher fluence requirements at 351-nm than these prior lasers. Thus the few small (□100μm) refractory inclusions present in conventional CVD fused silica is no longer acceptable for transmissive optics in the 351-nm section of the laser [40]. (Note, however, the conventional CVD SiO_2 material performs adequately [does not damage] in the 1053 nm section of the NIF laser.) Fortunately, significant investment has been made by suppliers of fused silica for lithographic applications such that this material can now be manufactured in large sizes (>40 × 40 cm^2) with high optical homogeneity and free of refractory inclusions [41,42].

III. Laser-induced-damage to optical glass surfaces

Recent pioneering work by Stuart et al. [22] has lead to a theory of surface damage to transparent dielectric materials that, when compared with recent experimental results from other authors [23,24], accurately predicts the surface damage threshold for silicate glasses over approximately five orders-of-magnitude in pulse length (Fig. 1). In the long-pulse region ($t_p > 20$ ps) laser damage is characterized by linear absorption and heating of conduction band electrons followed by transfer of the energy to the lattice; this in turn leads to heating and phase changes (melting and vaporization) of the material. In this region the onset of damage follows the well-known $\tau^{1/2}$ pulse-length behavior characteristic of thermal diffusion through the material. At shorter pulse lengths ($t_p \lesssim 20$ ps) the onset of damage departs from a $t_p^{1/2}$ dependence and can be theoretically described by the generation of free electrons via multiphoton ionization that are then rapidly heated in the oscillating electric field leading to collisional (avalanche) ionization. The electron heating is so fast that the energy transfer rate to the lattice is slow thereby generating rapid plasma formation and material ablation [22-24]; as a consequence there is negligible collateral damage outside of the ablation region. At 1000nm and pulse lengths below about 30 fs, multiphoton ionization alone provides the critical electron density needed for energy absorption from the beam and the resulting material ablation.

Laser induced surface damage is dominated by nano- to micro-scale absorbing defects and impurities for megajoule lasers with ns-pulselengths lasers (such as the NIF). Therefore, a number of recent studies have focused on quantifying the onset and extent of laser damage by various surface defects as well as seeking ways to reduce or eliminate them. For example, several studies have sought to quantify the impact of surface scratches and other visible surface defects on laser fluence limits [43,44]. In addition, work by Norton et al. has focused on understanding damage initiation and growth on SiO_2 surfaces visibly free of defects [45]. Norton et al. showed that initial damage sites are usually less than 50 μm in diameter and remain small at fluences less than about 5 J/cm^2 at 3-ns and 351 nm (Fig. 2). At fluences in excess of 5 J/cm^2, the initial damage sites grow exponentially with number of shots (Fig. 3) as characterized by the equation [45]:

$$D_N = D_o \exp(\alpha(F)N) \qquad (1)$$

where D_o is the initial damage spot size and D_N is the damage size after N laser shots at fluence, F (J/cm^2). The parameter $\alpha(F)$ is the exponential growth factor and at a 3-ns pulse length is linearly dependent on the fluence as given by [44]:

$$\alpha(F) = m \bullet F \ ; \ F \gtrsim 5.0 \text{ J/cm}^2 \qquad (2)$$

where m has been experimentally determined to be 0.04 ± 0.01 per shot per J/cm^2. Laser damage at visible defects (e.g. scratches) has also been shown to grow according to equation 1.

The proposed initiation source for SiO_2 surface damage is residual nano-scale contaminant particles left from the polishing process [46,47]. A highly schematic diagram of how these contaminant particles might become imbedded in or near the surface is depicted in Fig. 3. The diagram schematically illustrates how the surface roughness and subsurface fracture of an optical glass surface evolves during the different stages in the polishing process. The original ground glass surface has fractures that extend beneath the surface. Various approximation methods have been developed by master opticians for estimating the depth of sub-surface damage due to the grinding process (see for example [48]). Recently, researchers at the University of Rochester have sought to put this on a more scientific footing [49-53]. For example, Randi et al. [49] have reported an upper and lower bound to the depth of subsurface damage based on the size of the abrasive particle:

$$0.3L^{0.68} < d_f < 2L^{0.85} \qquad (3)$$

where d_f is the depth of subsurface damage (μm) and L is the abrasive size (μm); this is valid for abrasive sizes less than 100 μm. Furthermore these authors state that for optical glasses, the depth of subsurface damage is approximately the same as the peak-to-valley surface roughness and that all fractures are within a depth of twice the peak-to-valley roughness [49]. The "polishing" process uses finer abrasives to remove the layer containing sub-surface damage which in turn generate their own characteristic (but smaller) sub-surface fracture [48-55]. The goal is to essentially remove all sub-surface fractures by the time the surface achieves the final polished state (Fig. 3).

Due to the purely statistical nature of subsurface damage one has a lower probability of achieving completely "fracture-free" surfaces as the surface area increases. Therefore, for large laser systems with very large optical surface areas (hundreds of square meters), some level of residual subsurface damage almost certainly will be present. Sheehan et al. [56] have shown that the extent of residual subsurface damage can be visually detected by first etching the surface using a mild HF solution and then side-lighting the edges of the optic. Most of the white light that enters through the edges propagates through the optic by total internal reflection. The light scatters strongly at sub-surface defects and is readily detected by visual and/or microscopic examination.

One major source of nano-scale contamination that leads to laser damage is the residual grinding and polishing materials left in sub-surface fractures. The supernate liquid of the slurry enters the void space of the small surface fractures generated during grinding and polishing (Fig. 4). After finishing these fractures will ultimately "dry-out" leaving behind nano-scale debris. The size of the debris can be estimated from the measured dissolved-solids content of the polishing slurry (several weight percent) and the estimated volume of a residual fracture. In addition, it is well known that CeO_2, one of the most commonly used polishing compounds, interacts chemically with the glass surface, enhancing the material removal process [54,55,57,58]. Sheehan et al. have reported laser damage pits on the surface of CeO_2 polished fused silica irradiated at 351-nm [56]. They propose that residual CeO_2 nano-particles produce the damage pits that occur at such a high density as to give the damaged surface a hazy appearance. Also, more recently Wall et al. [59] have reported finding material from the polishing steps imbedded in the optic surface. In addition, analysis of the HF solution used to etch the glass shows the presence of residual polishing related contaminants [9,46,60].

IV. Improving the laser damage resistance of polished surfaces

Residual contaminants and sub-surface defects from the polishing process are generally ubiquitous. This has recently led researchers to propose that one must accept some level of contamination and defects from the conventional finishing processes and then employ a series of post-processing steps to reduce and ultimately eliminate laser-induced damage from these defects [9,19,25,61,62].

One post-processing step that has given a significant reduction in subsurface damage and defects is the newly developed magnetorheological finishing (MRF) process [9]. In contrast to conventional polishing which relies on a normal load applied to an abrasive particle to cause material removal, MRF uses a shear force [11,55]. In brief, the process makes use of a slurry containing ferromagnetic particles to create a synthetic polishing surface when the slurry passes into a magnetic field. The slurry also contains standard polishing components (e.g. CeO_2, diamond) [55] that drives the material removal process. The MRF process is described in detail elsewhere [11,55,63-67] and is commercially available in a suite of custom machine tools [68].

Menapace et al. have reported [9] a dramatic reduction in visible subsurface damage (after etching) for fused silica optics first finished by conventional means and then treated with an MRF post-processing step (Fig. 5). They report however, that the damage defect density at 351-nm of the MRF polished SiO_2 surface is improved only after the surface also has been acid etched (HF) to remove any residual ferromagnetic (iron carbonyl) contaminants from

the MRF process. This suggests that the MRF shear-force material removal process may not completely eliminate subsurface damage but rather simply reduces the depth and density of such fractures.

A second post-processing step for reducing defects that can be applied after the MRF process is "laser conditioning" [9,61]. Laser conditioning refers to the process of raster-scanning the full optic surface at incrementally higher laser fluences up to a point just below the region for damage onset. The conditioning process was first developed for improving the damage resistance of multilayer dielectric coatings [69] and potassium-dihydrogen-phosphate (KDP) crystals [70,71]. In the case of multi-layer coatings it has been shown that conditioning ejects loosely-held coating defects at low laser fluences such that collateral damage is avoided [72-74]. If the coatings are not conditioned then the collateral damage due to exposing these defects at a high fluence renders the coating unusable. It is probable that laser conditioning of a polished glass surface achieves the same effect as with a multilayer coating; it gently removes surface or near-surface defects with negligible collateral damage (i.e. does not produce damage that grows at high fluence). It is clear that one area of fruitful research is to develop a more fundamental understanding of the laser conditioning process on polished glass surfaces. Menapace et al. have shown that laser conditioning a MRF-polished, acid-etched surface at fluences up to 8 J/cm^2 at 351-nm and 7.5-ns can further reduce the residual surface damage density by up to 1000-fold [9].

Although MRF post-processing followed by acid-etching and then laser conditioning can reduce the damage density dramatically, these steps do not entirely eliminate all damage. Recently, Brusasco et al. [62] have demonstrated a method for initiating and then repairing any residual damage sites left on an optic surface (Fig. 6). The initiation is accomplished by simply scanning the optic at high fluence with a small diameter beam from a commercial laser operating at 351 nm. These initiated damage sites are then "repaired" by ablatively removing the damaged material via a timed exposure to the 10.6-μm output from a CW CO_2 laser [62] (Fig. 7). Subsequent testing shows that these "repaired" sites do not experience further damage or damage growth. Also the ablated sites are small enough that beam modulation due to diffraction from the site has a negligible impact on the down-stream optics.

It is clear that further research leading to a better understanding of polishing processes of glass surfaces will continue to benefit development of optical glasses for high power lasers. As one example, the work by Ito et al. [75,76] to develop a "less brittle" glass may lead to an improved understanding of glass composition/structure effects on surface material removal processes that could lead to even lower subsurface damage. The combination of

such glass composition work combined with research relating glass chemical and physical properties and slurry chemistry to material removal, such as the work at the University of Rochester [50-53], should dramatically advance not only our understanding of optical glass polishing but also laser-induced damage.

V. Summary and Conclusions

Laser induced damage to optical glasses continues to be a challenge in the operation of increasingly more powerful lasers. This problem requires scientific advances in many fields related to optical glasses, from improved theoretical descriptions of laser material interactions, to improved glass finishing methods, to an improved understanding of the material removal mechanism and corresponding generation of subsurface damage during optical finishing.

Laser induced damage to optical glasses can be categorized into either bulk (interior) or surface damage. Bulk damage is largely driven by the presence of metal or ceramic refractory inclusions that remain in the glass. Inclusion-free fused silica and metaphosphate laser glasses can now be made using recently developed melting and forming processes. Unfortunately these are the only glasses available for high-peak-power laser applications; new methods for preparing other optical glass types free of inclusions is a fruitful area of research.

One recent novel approach for dealing with surface initiated laser damage is to assume that one can not entirely rid the glass surface of all potential damage sites during polishing. Instead a series of post-polishing steps are used to reduce the number of sites and then finally the remaining sites are gently initiated and ablatively removed using CO_2 (10-μm) laser processing.

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Figure Captions

Figure 1. Comparison of predicted versus measured fluence for laser induced damage to polished SiO_2 at 1053 (•) and 526 nm (♦)[22] and to a barium alumino-borosilicate glass at 780nm (□) [24]. The theory (—) is from Stuart et al.; note the two limiting mechanism: multi-photon ionization (---) and thermal diffusion (...)

Figure 2. (a) Exponential growth of a 225-μm laser-induced surface damage site at 351-nm (10.5 ± 1.5-ns) with number of laser shots at a fluence of 6.5 (±1.0) J/cm^2 [45]. The solid line is a fit of equation 1 to the data. (b) Growth coefficient ($\alpha(F)$) for laser-induced surface damage to polished fused silica surfaces as a function of laser fluence[45]. The solid line is given by equation 2

Figure 3. Schematic diagram of the successive stages of material removal used to achieve a highly polished glass surface. Each material removal step generates subsurface fractures that extend below the surface with some characteristic depth distribution.

Figure 4. Schematic representation of (a) fluid from the finishing slurry entering a sub-surface fracture that (b) eventually "dries" to leave behind nano-scale contaminants.

Figure 5. Residual sub-surface defect sites for a polished SiO_2 surface after (a) conventional polishing and (b) MRF polishing. The sites are made visible by first etching the glass surface and then side-lighting the optic as described in the text. Note the dramatically lower density of defects on the MRF finished surface.

Figure 6. (a) Laser-induced damage site on the surface of fused silica and (b) corresponding depth profile before CO_2 laser treatment. Photograph (c) of the same site, and (d) respective depth profile after CO_2 laser ablation of the damaged region [62]. Note the ablation process leaves a non-damaging, smooth, gaussian-shaped pit.

Figure 7. Schematic diagram of process used to (a) initiate and then (b) repair laser-induced damage on a polished fused silica optic [19,62].

Figure 1.

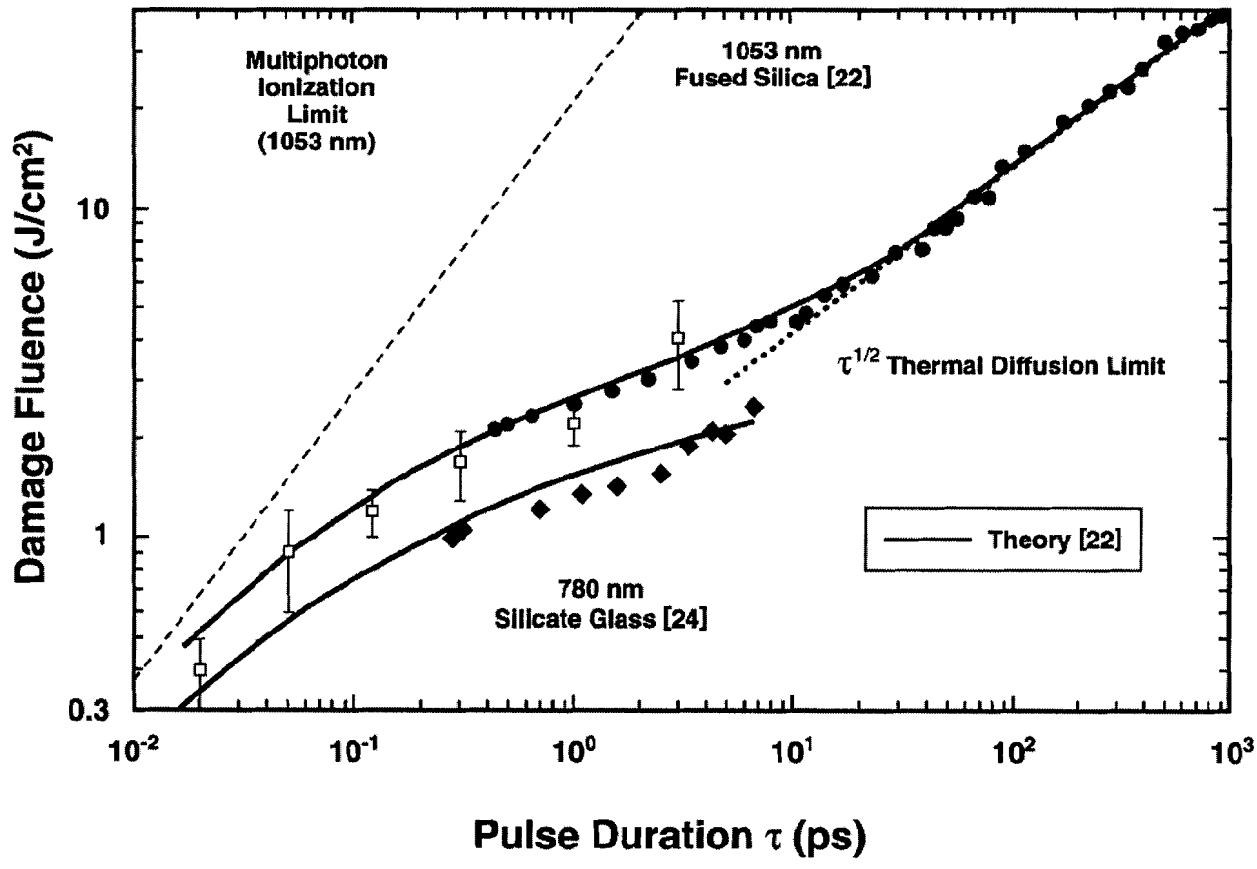

Multiphoton
Ionization
Limit
(1053 nm)
1053 nm
Fused Silica [22]
τ^1/2 Thermal Diffusion Limit
Theory [22]
780 nm
Silicate Glass [24]
Damage Fluence (J/cm²)
10
1
0.3
10^-2
10^-1
10^0
10^1
10^2
10^3
Pulse Duration τ (ps)

Figure 2

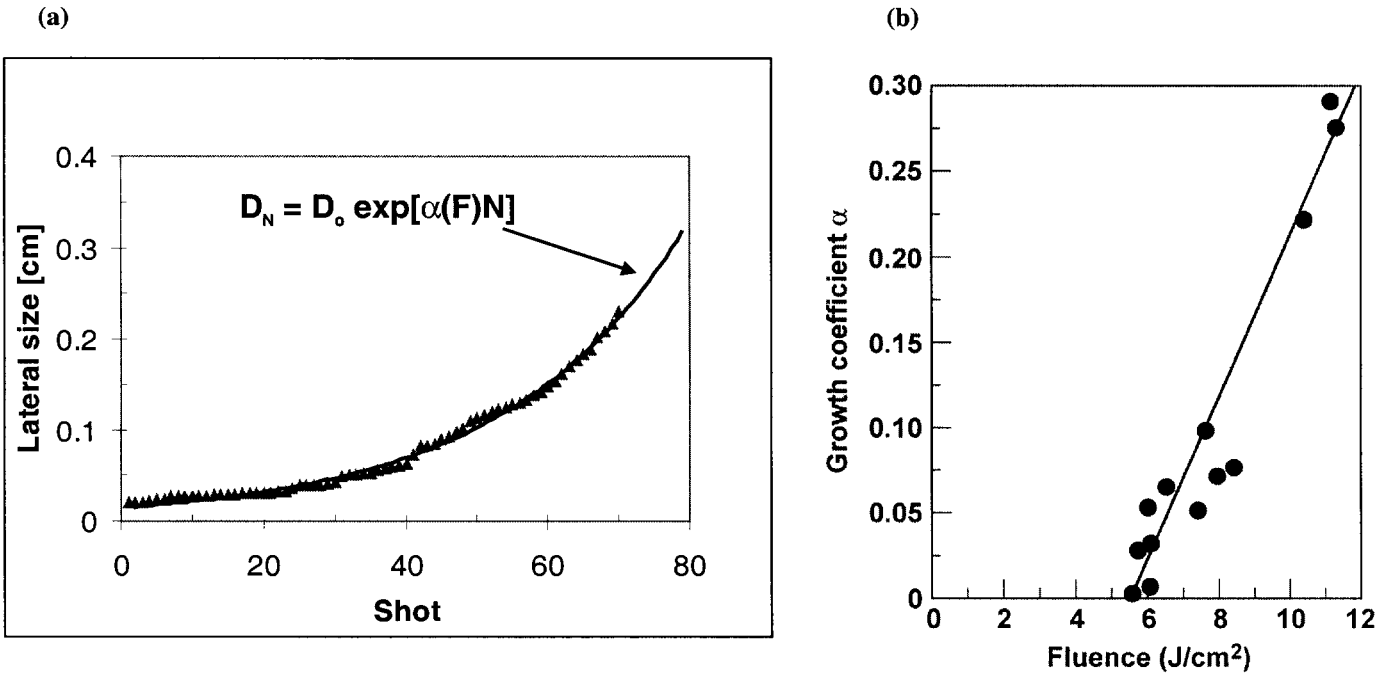

(a)
(b)
$D_N = D_o \exp[\alpha(F)N]$
Lateral size [cm]
Shot
Growth coefficient α
Fluence (J/cm²)

Figure 3.

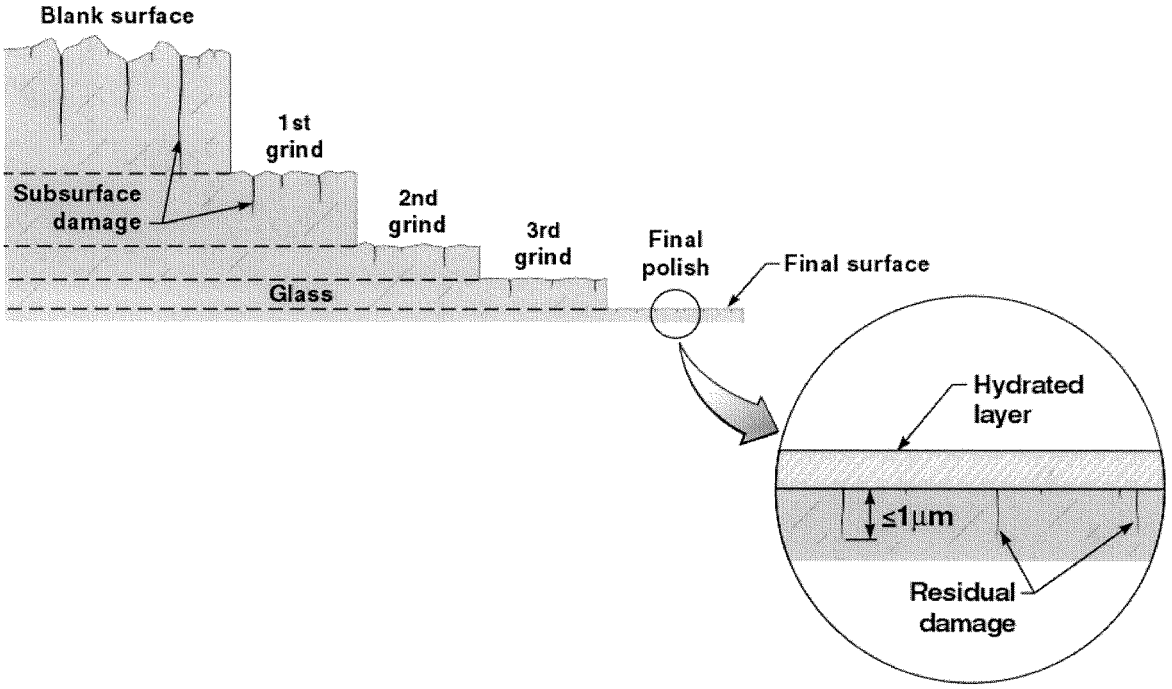

Blank surface
1st grind
Subsurface damage
2nd grind
3rd grind
Final polish
Final surface
Glass
Hydrated layer
≤1μm
Residual damage

Figure 4.

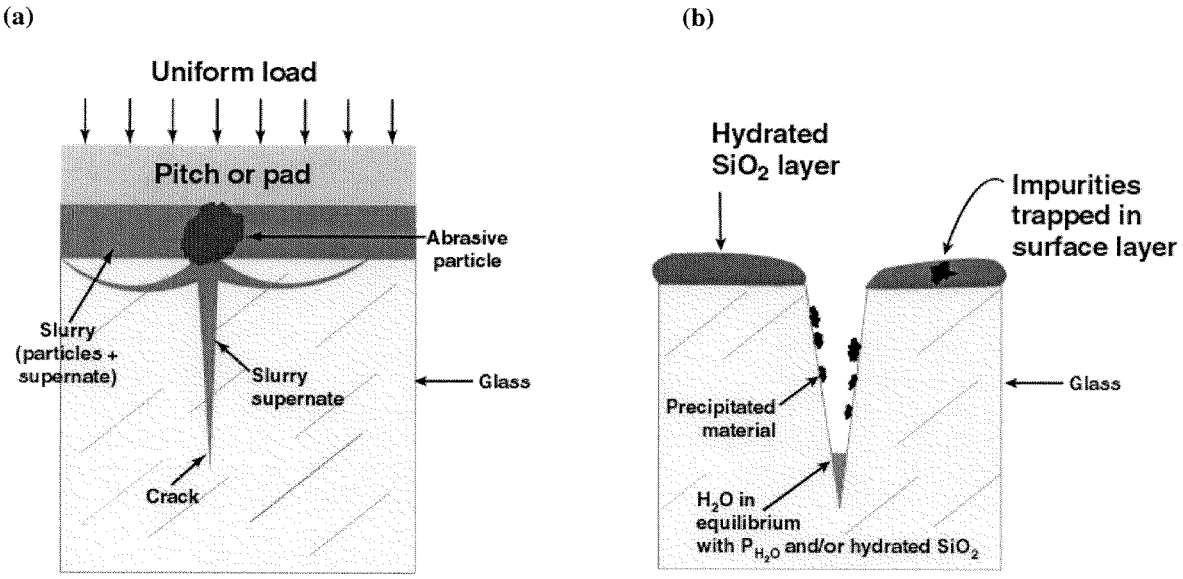
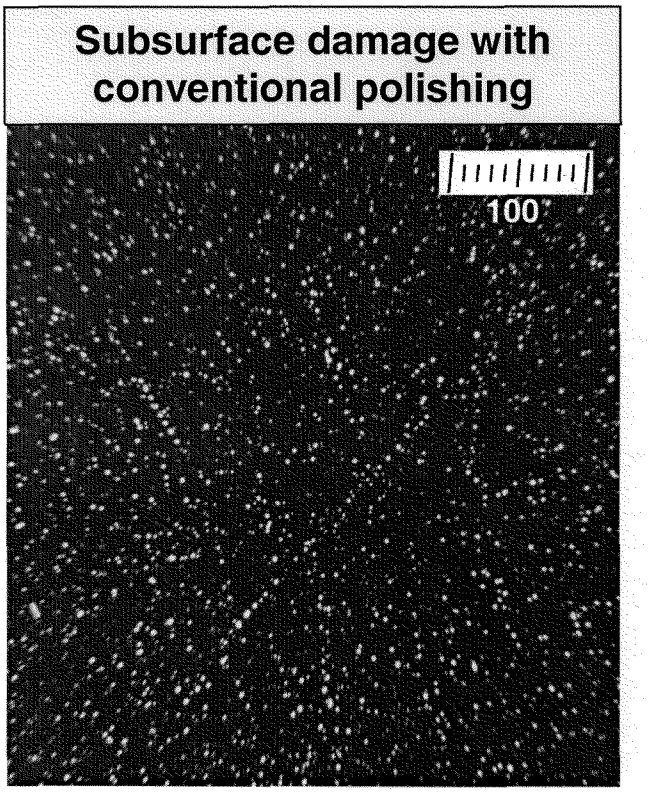

(a)
Uniform load
Pitch or pad
Abrasive particle
Slurry (particles + supernate)
Slurry supernate
Glass
Crack
(b)
Hydrated SiO_2 layer
Impurities trapped in surface layer
Glass
Precipitated material
H_2O in equilibrium with P_{H_2O} and/or hydrated SiO_2

Figure 5.

(a)


Subsurface damage with conventional polishing
100

(b)

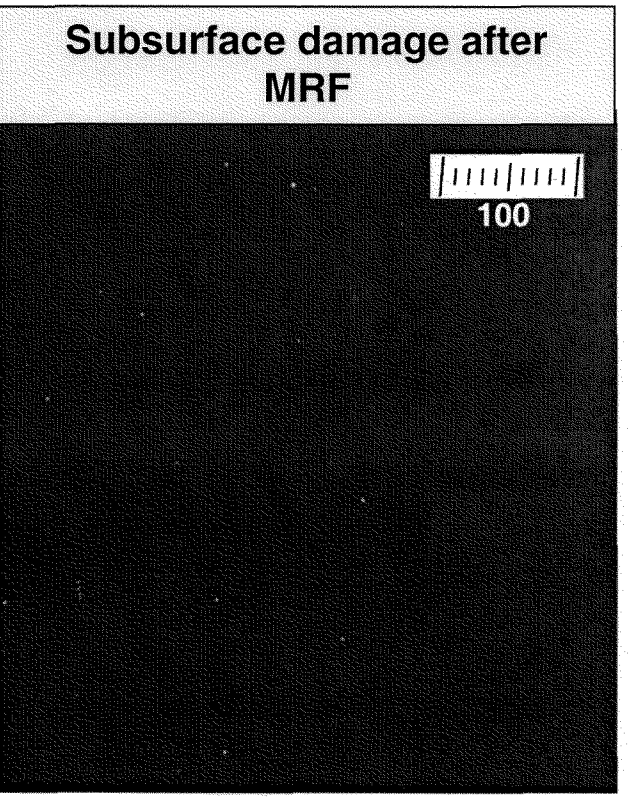

Subsurface damage after MRF
100

Figure 6.

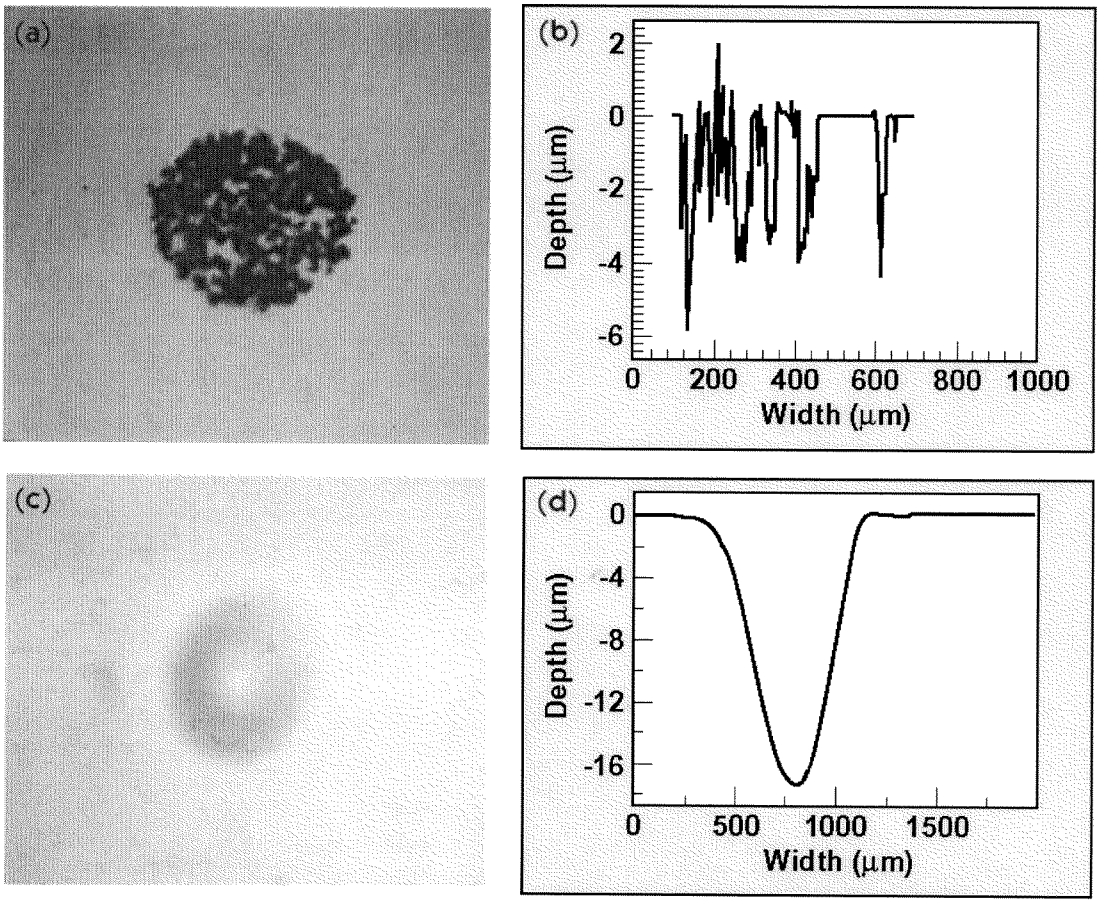
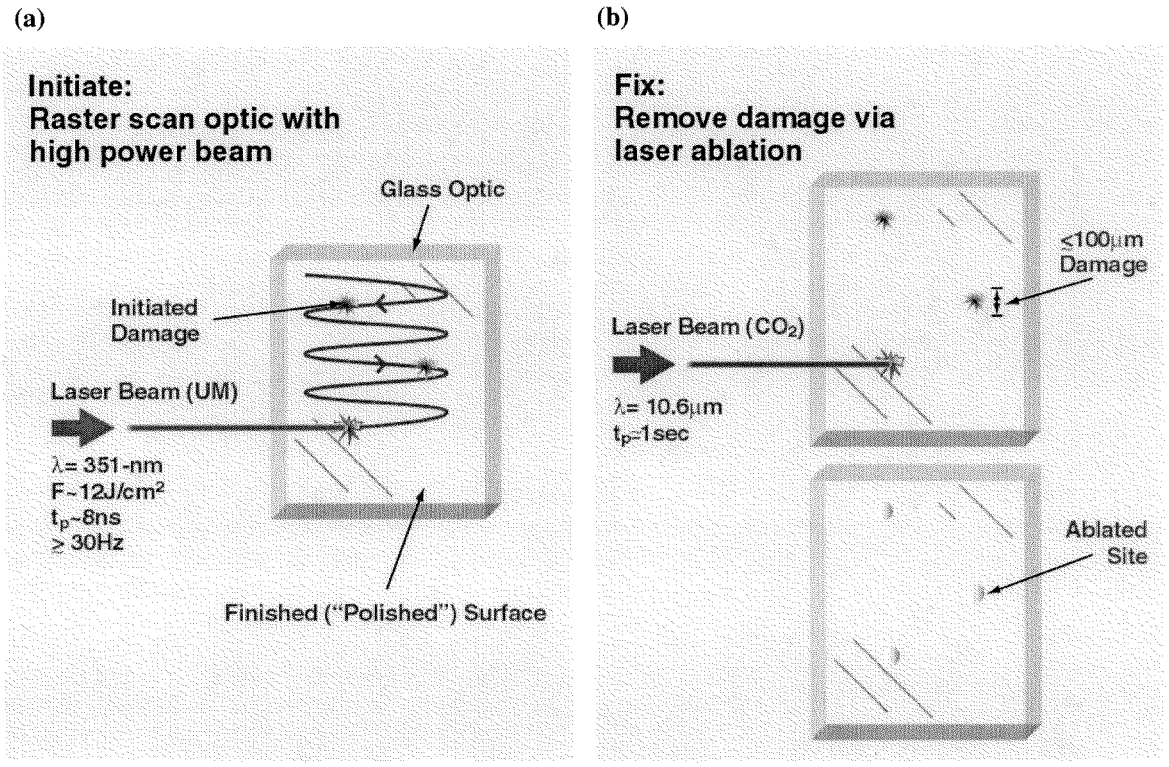

(a)
(b)
Depth (μm)
2
0
-2
-4
-6
0
200
400
600
800
1000
Width (μm)
(c)
(d)
Depth (μm)
0
-4
-8
-12
-16
0
500
1000
1500
Width (μm)

Figure 7.


(a)
Initiate:
Raster scan optic with
high power beam
Glass Optic
Initiated
Damage
Laser Beam (UM)
λ= 351-nm
F~12J/cm²
tp~8ns
≥ 30Hz
Finished ("Polished") Surface
(b)
Fix:
Remove damage via
laser ablation
≤100μm
Damage
Laser Beam (CO2)
λ= 10.6μm
tp=1sec
Ablated
Site