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# Advanced surface processing of NLO borate crystals for UV generation

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Abstract Recent advances in NLO Borate Crystals for UV Generation are reviewed with the particular emphasis on the technique to improve the life time of UV optics. The laser-damage resistance of CLBO and fused silica surfaces was successfully improved after removing polishing compound by ion beam etching. The polishing compound embedded in the CLBO and fused silica surfaces were to a depth of less than 100 nm. We were able to remove polishing compound without degrading the surface condition when the applied ion beam voltage was less than 200 V. The laser-induced surface damage threshold of CLBO was improved up to 15 J/cm$^2$ (wavelength: 355 nm, pulse width: 0.85 ns) as compared with that of the as-polished surface (11 J/cm$^2$). The laser-induced surface damage of fused silica also increased from 7.5 J/cm$^2$ to 15 J/cm$^2$. For the irradiation of a 266 nm high-intensity and high-repetition laser light, the surface lifetime of CLBO and fused silica could be more doubled compared with that of the as-polished surface.

## 1. Introduction

Considerable attention has been paid over the years to the improvement of laser-induced damage threshold (LIDT) on UV optics. The damage of optics, which is typically initiated in the as-polished surface, often limits the UV output power of lasers. The laser-induced surface damage has been linked to energy-absorbing defects such as surface contamination and surface scratches. These defects occur during the grinding and polishing processes used to finish the optical surfaces. Surface scratches are eliminated and/or minimized in practice by using a controlled sequence of successively gentler grinding and polishing steps. On the other hand, contamination, that from polishing compound can not be prevented in conventional mechanical polishing process. Ceria ($CeO_2$) and zirconia ($ZrO_2$), are favorites polishing compound, but are strong absorbers at shorter wavelength than 355 nm. On the as-polished surface of optics, polishing compound are embedding inside the near-surface region. To attempt to increase the laser damage resistivity of UV optics, these polishing compound lowering the laser-induced damage threshold must remove from the surface. We have developed the Ar ion beam etching process in order to etch off the polishing compound from the optics surface [1-3]. A nonlinear optical (NLO) crystal $CsLiB_6O_{10}$ (CLBO) using for the fourth (266 nm) and fifth (213 nm) harmonic generation of Nd : YAG lasers [4-8] and fused silica surfaces were used as UV optics. In this study, the ion beam etching of the optical surface and effects of polishing compound removal on surface damage durability were investigated.

## 2. Experimental

### 2.1. *Surface etching procedures*

The investigations were performed on $ZrO_2$ polished (100) surfaces of CLBO samples with dimensions of $10\times10\times3$ mm$^3$, provided by Kogakugiken Co., Ltd., and $CeO_2$ polished UV-grade 40-mm diameter fused silica samples. These mechanically polished sample surfaces are contaminated with polishing contaminants in the polishing slurry. The polishing compound was detected in the sample surfaces to a depth of 100 nm (CLBO was 60 nm, fused silica was 50~60 nm). The quantity of polishing compound decreases with increasing depth. A neutralized ion beam produced by a Kaufman ion source was used to remove the polishing compound. The experimental arrangement is shown in Fig. 1. The base pressure of the chamber was $5\times10^{-6}$ Torr or lower. Argon was used as a sputtering gas and was kept at a constant pressure of about $3\times10^{-4}$ Torr by means of a mass-flow meter. The distance from the sample to the ion source was approximately 25 cm. The samples were fixed on the etching tool and continuously rotated at an incidence angle of 45°. After surface etching, the etched-surface qualities were characterized. Scanning atomic force microscopy

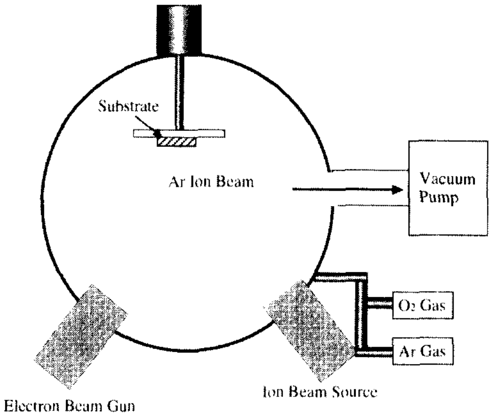


Fig. 1. Ion beam etching system.

(AFM) was used to investigate the surface roughness before and after etching. Surface damage due to ion bombardment was observed using reflective X-ray topograph. Laser-induced damage threshold (LIDT) testing was performed on each etched sample to investigate the effects of the ion beam etching.

## 3. Results and discussion

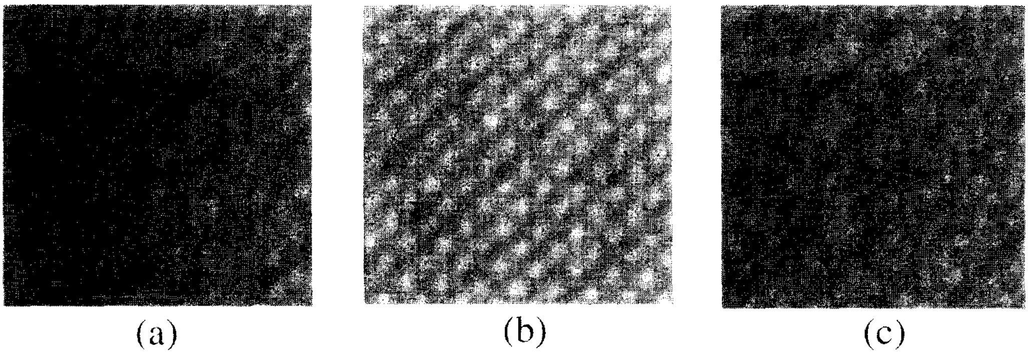
### *3.1. Dependence of surface quality on etching conditions*

Polishing compound embedded in the sample surfaces was removed by using ion-beam etching. The surface condition will be degraded if the ion voltage is too intense or applied for too long. In particular, the increase in surface roughness and occurrence of surface damage due to ion bombardment are serious problems for the laser-induced surface damage of high-power optics. Increasing in surface roughness locally enhances the electric field strength and scattering loss [9]. The polishing compound must therefore be removed without degrading surface conditions. The quality of the surface condition treated with ion beam depends on the applied ion-beam voltage. In this study, samples were etched with ion beam voltages of 200, 300 and 400 V for 1 hour. Table 1, shows the applied ion beam voltage related to surface roughness at an argon gas pressure of $3\times10^{-4}$ Torr. At a low ion beam voltage of 200 V, the surface roughness of CLBO did not increase. However, the surface roughness increases considerably at an applied ion-beam voltage of 400 V. The magnitude of increased surface roughness was 2.66 nm rms. However, there is no relation between applied ion-beam voltage and surface roughness for fused silica. The as-polished fused silica surface roughness was about 0.52 nm rms. The surface roughness when the surface was etched with an ion-beam voltage of 400 V was the same as the as-polished surface roughness.

Table 1
Relationship of surface roughness with applied ion-beam voltage at an argon gas pressure of 4×10-4 Torr

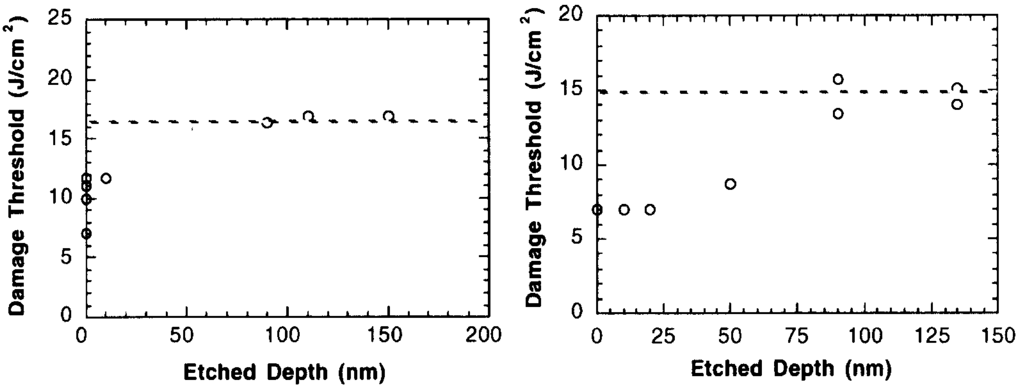
| Ion beam voltage (V) | CLBO | | Fused silica | |
|---|---|---|---|---|
| | RMS (nm) | Mean height (nm) | RMS (nm) | Mean height (nm) |
| 0 (as-polished) | 0.60 | 2.20 | 0.52 | 2.42 |
| 200 | 0.52 | 2.14 | 0.33 | 1.69 |
| 400 | 3.26 | 10.85 | 0.44 | 1.99 |

There is a general understanding that if the energy of the bombardment ions is increased sufficiently, some damage to the etched surface will occur. The



(a) (b) (c)

Fig. 2. Reflective X-ray topographs for (100) surfaces. (a) as-polished. (b) ion beam etched at 200 V. (c) ion-beam etched at 400 V.



(a) (b)

Fig. 3. Effect of ion beam etching on single-shot entrance surface LIDT at 355 nm. (a) CLBO. (b) fused silica.

surface damage as a result of ion-beam etching was investigated for the CLBO surface. Figures 2 (a), (b) and (c) show the results of a reflective X-ray topography for the (100) surfaces of CLBO. Surface damage was not observed on ion beam etched surfaces (b) and (c) as compared with than that of as-polished surface (a). These results indicate that the surface condition depends on the ion beam voltage, and a lower ion beam voltage can remove polishing compound without degrading the surface. For CLBO crystal, degradation of the surface condition was not observed for ion beam voltages of up to 200 V.

### *3.2. Effect of ion beam etching on surface damage*

CLBO and fused silica samples were etched at various etching depths with an ion-beam voltage of 200 V. Surface damage of the etched samples was investigated as a function of an etching depth for two laser sources. One is single-shot laser and another is high-repetition laser. For the shingle-shot test, the 355 nm laser beam was focused on the etched entrance surface by a lens with a focal length of 100 mm. The focused laser beam had a pulse width of 0.85 ns and a smooth Gaussian shape with a spot diameter at $1/e^2$ peak intensity of 300 μm. A Nomarski microscope at 200X was used to detect damage. The LIDT was defined to be that fluence below which no damage occurs. The results of LIDT for CLBO and fused silica are shown in Fig. 3 (a) and (b). The as-polished surface of CLBO had damage thresholds at around 10 J/cm$^2$. The LIDTs increase with ion beam etching depth in all samples. For the surfaces etched more than 60 nm, ion-beam etching improves laser-damage resistance up to 1.6 times. The LIDT of those etched more than 60 nm was about 16 J/cm$^2$, compared to 10 J/cm$^2$ of the as-polished sample. In the fused silica surface, we also found the improvement of surface LIDT up to 15 J/cm$^2$ as compared with that of the as-polished surface of 7.5 J/cm$^2$.

For the high-repetition laser, the lifetime testing was performed using a 266 nm of Nd : YAG laser operating with a repetition rate of 3 kHz and a pulse width of 76 ns. In this experiments, irradiated spot size was accuracy measured using knife-edge test. To measure the surface lifetime acceleratingly, the power density on irradiated spots was adjusted to about 30 MW/cm$^2$. Surface damage was indicated by a decrease of the transmitted laser power. Figure 4, shows the results of accelerated lifetime tests of the exit surface of CLBO at various etching depths. For the as-polished sample,

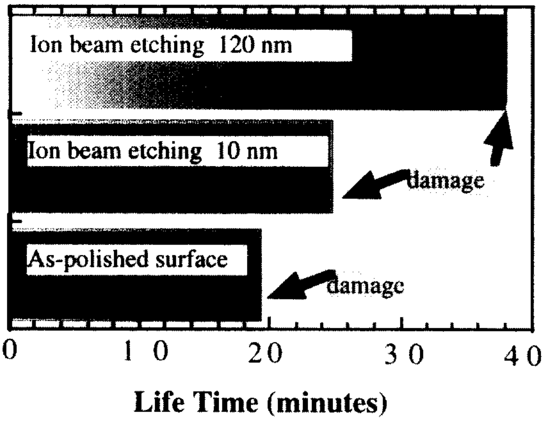


Fig. 4. Lifetime of CLBO surfaces with laser damage for the irradiating of 266 nm laser light (30 MW/cm$^2$).

surface lifetime was up to 19 minutes. For the sample etched to 10 nm, surface lifetime was increased to 25 minutes. Finally, the sample etched to 120 nm indicates an improved lifetime of 40 minutes. Thus, surface durability was improved by a factor of two, compared with that of the as-polished surface. From these results, it is clear that pretreating an optical surface by ion-beam etching can improve laser-damage resistance.

## 4. Conclusions

In conclusion, the effect of ion-beam etching on surface damage in CLBO and fused silica has been studied. Mechanically polished surfaces are contaminated with polishing contaminants in the polishing slurry. Polishing compound was detected in the as-polished surface to a depth of about 100 nm (CLBO 60 nm, fused silica less than 100 nm), and is difficult to remove by traditional cleaning techniques. Ion-beam etching was able to remove polishing compound embedded inside as-polished surfaces. The surface degradation resulting from surface etching depends on the applied ion-beam voltage. No detectable damage was observed when the applied ion-beam voltage was less than 200 V. Laser-damage resistance of etched surfaces was tested for LIDT (355 nm, 0.85 ns) and surface lifetime (266 nm, 76 ns, 3 kHz). The laser-damage resistance of fused silica and CLBO was improved with ion-beam etching for almost all samples. The LIDT of fused silica increased from 7 J/cm$^2$ to 15 J/cm$^2$ at more than 90 nm ion-beam etching. The improvement of laser-damage resistance for CLBO was the same as that of the fused silica. The LIDT of CLBO was improved up to 15 J/cm$^2$ by ion-beam etching. The surface lifetime for high-repetition, high-power-density irradiation at 266 nm was also increased by more than 2 times compared with that of the as-polished surface. This technique is very useful in fabricating high laser-damage resistant optical surfaces for high-power UV lasers.

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