Direct-write waveguides and structural modification by femtosecond laser pulses

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ABSTRACT

Embedded waveguides and their optical properties in bulk silicate glasses fabricated by femtosecond (fs) laser pulses (800 nm, <120 fs, at 1 kHz) are reported. Experimental results show that there is a narrow operating window for our system to produce low loss waveguides. An angular dependence of light transmission measured between two crossed polarizers on these laser-modified regions suggests that these regions possess an optical birefringent property. Furthermore, the optical axes of laser-induced birefringence can be controlled by the polarization direction of the fs laser. Permanent optical birefringence induced by the fs laser pulses can be produced in amorphous silica, and borosilicate glass. Raman spectroscopy of the modified glass shows a densification and reconstruction of silica network in the glass. Results show that the amount of laser-induced birefringence depends on pulse energy level and the number of accumulated pulses. Mechanisms that contribute to the observed laser induced birefringence behavior are discussed.

Keywords: Femtosecond, Raman spectra, waveguides, birefringence

1. INTRODUCTION

When an intense femtosecond (fs) pulsed laser is focused inside a bulk transparent material, the intensity in the local volume can become high enough to cause absorption through nonlinear processes, leading to optical breakdown or photo-damage. Slightly below the optical breakdown threshold, this nonlinear, multi-photon absorption process can locally alter the structure of bulk transparent material and cause refractive index change. Previous experimental evidence has shown two possible mechanisms involved in the bulk refractive index modification in silica-based glasses, which are (1) localized densification of the glass, (2) defect creation such as Si E', non-bridging oxygen hole centers, and peroxy-radicals. Similar to other high-energy photons or particles, the enormous power density of a fs pulse can easily create defects in glass. These defects and color centers can produce a significant change in refractive index. However, most of these defects can be easily removed during a high temperature heat treatment (>350°C). Because the refractive index change by the fs laser pulse in fused quartz cannot be removed by the traditional thermal treatment, the mechanism contributing to the permanent change of refractive index in glass favors the localized densification. Depending on the peak power of a fs laser pulse, local densification can be achieved by bulk heating with a high repetition rate and structural rearrangement in the silica network. In this study, we examine the effect of refractive index modification as a function of glass composition. Different optical grade glasses were selected for this study. Special emphasis is placed on the amount of glass modifiers present in the different glass compositions, and the effect of their glass transition temperature on the local structural change. The effect of processing parameters such as pulse energy level, and accumulated fluence on the optical property change will be reported. The local structural modification is characterized by Raman spectroscopy. Results will be compared with optical loss measurements and microstructure changes on photo-damaged areas to elucidate the possible mechanism that create the unusual optical property.

In addition to the fundamental interest in the processes that lead to permanent refractive index change, there is great interest in using this approach to create three-dimensional waveguides and integrated optics in bulk glass. Recent work has demonstrated that good waveguide properties can be achieved by moving the focus spot of fs laser pulses inside the bulk glass. In this paper, we will summarize the effect of processing conditions on the waveguide properties and report an unusual fs laser induced birefringence in an optically isotropic glass. Several earlier reports indicate that when a fs laser focuses into a bulk glass optic anisotropy can be created. However, systematic study the effect of processing conditions and quantitative measurements on the amount of birefringence induced by the laser pulses were not evidence, and will be the subject of this investigation. The capability to create optical birefringence in amorphous
silicon dioxide material provides a new pathway to control the polarization of light in silicon dioxide optical waveguides and will have significant impacts on integrated optics.

2. EXPERIMENTAL PROCEDURE

The laser used in this experiment is a regeneratively amplified Ti:Sapphire femtosecond pulsed laser (Spectra-Physics, Hurricane). The fs laser pulses have a nominal 123-fs pulse-width (FWHM) and 1 kHz pulse repetition rate. The energy of the pulse can be varied by the combination of a half wave plate and a thin film polarizer. Further energy reduction is accomplished by the use of neutral density filters to avoid photo-damage to the polished glass plates. The average laser power used in this investigation, without creating photo-damage to the glass, is between 450 µW and 900 µW which corresponds to a pulse energy between 0.45 µJ and 0.9 µJ. The laser is focused to a 3 µm diameter spot inside the glass, using a long-working-distance microscope objective (Mitutoyo, 20X, NA = 0.40). The calculated peak intensity of the fs pulse is 9.8 ×10²⁰ W/m² for a pulse energy of 0.9µJ.

In this investigation, six different optical grade glasses were investigated. The composition, soft point (T_s), and glass transition temperature (T_g) of these glasses are tabulated in Table I. The fused quartz, CVD quartz, and Pyrex glass consist a high percentage of glass former and tend to form extensive three-dimensional networks of silica tetrahedral. As a result, these glasses have a higher glass transition temperature and soft point. Glasses having a large amount of glass modifiers, such as alkali and alkaline metal oxides, tend to form a more open structure and possess a lower glass transition temperature. For the laser-induced birefringence study, a thermally treated chemical vapor deposited (CVD) amorphous silica plate (Quartz International, Albuquerque, NM) was used in this investigation since there was no built-in birefringence observed before laser writing. The glass plate (polished on both surfaces) was mounted on a motorized x-y translation stage with linearly controlled motion along the z-direction to adjust the focus of the laser inside of glass. The laser was focused approximately 600 µm below the surface of the glass plates. In addition to these linear motion controls, the sample can also be rotated in the x-y plane with a rotational stage. In the writing process for the birefringence study, the polarization direction of the beam was fixed and the sample was rotated with respect to the polarization direction of the laser beam. By moving the laser focus spot perpendicular to the beam direction (20 µm/s) with an appropriate energy level (0.45 to 0.9 µJ per pulse) in side of bulk glass, optic waveguides can be created inside of bulk glass. The general quality of these waveguides was studied by a near-field and a far-field method.

The laser-induced local structural modification was analyzed, using the microscope accessory to the Raman spectrometer and the 458 nm laser line as an excitation source. A microscope objective focused the excitation laser into the glass and collected 180° backscattered light for dispersion and detection by the spectrometer. Collected light was dispersed by a triple monochromator and detected by a charge-coupled detector. The volume sampled using this technique approximates a cylinder of 1 µm diameter and 3-6 µm in depth, which is perpendicular to the propagation direction of the laser beam. After the laser writing, the samples were evaluated using a transmission microscope (Olympus BH-2). The illumination light is monochromatic light filtered from a halogen lamp. It has a peak wavelength at 671 nm and full width at half maximum bandwidth of 10 nm. The samples were put between two crossed polarizers on the rotation stage of the microscope. The angular rotation of the sample with respect to the polarizer was accurately measured by the stage goniometer. The intensities of the transmitted light through the samples were measured by using a CCD digital camera attached to the transmission microscope viewer. Optical transmission signals were captured using a National Instruments PCI-1424 digital image capture card. The CCD camera is operated in the linear regime. The maximum intensity of the transmitted light for the laser-modified region was determined by scanning the image array for the maximum value in each of ten images. The value was then averaged and recorded.

3. RESULTS AND DISCUSSION

3.1 Structure modification and nonlinear absorption

Because of the nonlinear absorption processes, femtosecond laser pulses can be focused deep inside of bulk glass and alter the local glass structure and create refractive index modification. If the intensity of the laser is below the threshold
level of multi-photon absorption, glass becomes almost transparent to the laser beam. Adversely, if the intensity of the beam is too high, it can easily create optic damage to the glass. Therefore, it is important to identify the proper processing space that can effectively change the refractive index without creating photo-damage in the bulk glass. Our experimental results show the threshold of pulse energy that can create a detectable waveguide pattern by the nonlinear absorption is above 0.2 µJ. Good quality waveguides can only be created in a very narrow processing window i.e., with pulse energy between 0.45 to 0.90 µJ. Above this energy level, stress birefringence and sometimes microcracks can be observed, indicating photo-damage to the bulk glass. The observation is consistent with data in the literature.5

Experimental results show that creating refractive index changes on the glass surface is extremely difficult, if not impossible. Attempts using different optics and power level to write a surface waveguide were unsuccessful. It is found that there required a minimum light penetration depth beyond which the change of refractive index by the fs laser pulses can be performed. Results show that a minimum depth of 15 µm is required for fused quartz. The probability for the multiphoton absorption directly from 800 nm wavelength photons is minute since the process requires stacking more than 5 photons to cross the band gap of fused quartz. It is believed that the absorption process comes from the absorption of two to three high energy photons created by the self phase modulation when an intense fs laser pulse passes through an optical media. As a result, there is a minimum penetration depth before enough photon energy can be absorbed by the self phase modulation and the multiphoton absorption processes for the local structural modification in bulk glass.

The observation of a narrow laser processing window and a minimum light penetration depth in creating the refractive index change in bulk glass confirms that the local glass structure modification is created by a nonlinear multiphoton absorption process.

3.2 Structure modification by the femtosecond laser pulses
3.2.1 Changes in the Raman spectra of laser-damaged region
Figure 1 shows the micro-Raman spectrum of bulk fused quartz and normalized spectrum of a laser-damaged region (0.1 mJ). Under a cross-polarized light condition, the damaged regions exhibit stress birefringence and are occasionally associated with microcracks. Without the normalization procedure, the intensity of the Raman bands from the laser-damaged region is consistently less than that from the bulk glass. This reduction in intensity indicates that the modified regions have structures of lower density where some glass has been partially replaced by voids. Furthermore, the normalized Raman spectra show an increase in the 490 and 605 cm\(^{-1}\) peaks in the damaged region, indicating an increase in the number of 4- and 3- membered ring structures\(^{13, 14}\) in the silica network. The formation of these 4- and 3- ring structures is sometimes associated with the creation of new quasi-surfaces at small voids in the silica-rich matrices.\(^{15}\) In addition, the Raman band peaking between 400 and 500 cm\(^{-1}\) is significantly narrower in the spectra from laser-modified regions compared to the bulk glass. A similar behavior is observed for the laser-modified region in Pyrex glass. The relative narrowness of the bands in the spectra indicates that the silica networks in these areas have a smaller distribution of ring sizes, with fewer large rings. This type of modification of the silica network is typically achieved by quick melting followed by fast re-solidification during the laser process. The relatively high melting points of fused quartz and Pyrex contributed to their fast re-solidification.

In contrast to the extensively three-dimensional networked silica tetrahedral in fused quartz (or CVD quartz) and Pyrex glass, the Raman spectra of glasses with high concentration of glass modifier (lead glasses, in Figure 2) show less effect in the changes of the Raman bands intensity from a laser-damaged region. However, a significant change in the Raman intensity can be observed below 500 cm\(^{-1}\), suggesting a change of the silica network in these glasses. In addition, the change of Raman intensity below 500 cm\(^{-1}\) for these glasses decreases with decreasing of \(T_g\). The smaller change in the Raman spectra can, therefore, be attributed to the lower melting point (and \(T_g\)) of these glasses, where the structural relaxation occurs much faster.

3.3.2 Changes in the Raman Spectra of laser-modified glass
Figure 3 summarizes the Raman spectra data of laser-modified regions from the Pyrex glass samples, where the laser energy is below photo-damage threshold. Results show that the overall reduction of the intensity of Raman bands becomes significantly less for the laser-modified region. Similar to the photo-damaged area, the small reduction of the intensities of Raman bands can be contributed to the creation of voids in the laser-modified areas. In addition, the 445 cm\(^{-1}\) band in the spectrum is shifted to high frequency, and a Raman band peaking near 603 cm\(^{-1}\) appears for laser-
modified regions. The upward shift in the 445 cm⁻¹ band indicates the formation of 4-membered silica rings, and the appearance of the 603 cm⁻¹ band indicates the formation of 3-membered silica rings. Furthermore, an enhanced Rayleigh scattering of the laser beam in the laser-modified regions during Raman study was observed, an indication of a difference in refractive index between modified and unmodified regions. The formation of 3- and 4- membered rings by the fs laser pulses, as well as a slight reduction in the overall intensity of the Raman bands are also observed in CVD quartz with laser energy below the photo-damage threshold. Detailed analysis show that the Raman feature for the 4-membered silica ring at 490 cm⁻¹ in the CVD quartz has moved up to 512 cm⁻¹ in the laser-modified regions (not shown), suggesting the newly formed 4-membered silica rings are under a compressive stress. In contrast to the photo-damaged area, the narrowing of the Raman band peaking between 400 and 500 cm⁻¹ is not observed in the laser-modified regions.

3.3 Embedded waveguides
With the knowledge of the effect of average laser power on structure modification of glass, direct-writing waveguides inside amorphous quartz was explored. Figure 4 shows the top view, near-field and far-field patterns of laser-modified waveguides above (0.1 mJ) and below photo-damage threshold (0.9 µJ) in thermally treated CVD quartz. The near-field and far-field patterns were obtained from the transmitted beams at 650 nm through 2 mm length of laser-modified regions. The near-field pattern of waveguides created below the damage threshold illustrates the propagation of a single LP01 mode through the waveguide inside of a bulk glass. The far-field pattern indicates that this transmitted mode possesses a nearly Gaussian profile. No far-field patterns can be obtained due to a significant transmission loss in the waveguides. The transmission loss determined from the top view is summarized in Figure 5. Above the photo-damage threshold (> 0.9 µJ), extensive light scattering is observed, which is consistent with the Raman analysis results where some glass has been partially replaced by voids. Results also show that below the photo-damage threshold, the transmission loss is almost negligible within 2 mm. The embedded waveguides in the amorphous quartz are preserved after a half hour thermal treatment at 650°C, indicating a permanent change of refractive index created by the fs laser pulses. These results demonstrate the potential for fabricating integrated optics by the fs laser pulses.

3.4 Laser-induced birefringence in optically isotropic glass
Figure 6 (a) illustrates the variation in transmitted light intensity through the laser-modified regions between two cross-polars. Each modified region was exposed to 60,000 pulses (1 minute dwell time) parallel to the beam direction with an average pulse energy of 0.9 µJ. The polarization direction of the laser beam was systematically rotated 10° from spot to spot, by rotating sample with respect to the polarization direction of the laser beam. The observation of an optical extinction for every 90° suggests that the laser-modified regions possess an optical birefringence property. Furthermore, the sinusoidal variation of the transmitted light intensity (see Figure 6 (b)) is consistent with the theoretical prediction of a birefringent material. Assuming that there is no significant losses from reflection and absorption from these laser-modified regions, the maximum change of birefringence (Δnₐ) induced by the laser is in the range of 0.002 to 0.004 (Figure 7), depending on the number of accumulated laser pulses. Results indicate that the laser-induced birefringence property in the bulk glass can be controlled by laser power level, accumulated exposure, and polarization direction of the writing laser. Details of the quantitative measurement of birefringence created by the fs laser pulse will be reported elsewhere.

Although the fundamental mechanisms that create the birefringence in the bulk glass by the fs laser irradiation is not clear yet, several observations in this study have helped to shed light on this issue. Results have shown that (1) the high energy linearly polarized laser pulse can induce birefringence in optically isotropic glasses, (2) the laser induced birefringence can be controlled by the polarization direction of the laser beam. The polarization dependence of birefringence immediately negates the possibility of a pure thermal effect generated by multiphoton absorption or plasma formation. If the thermal heating is a major contributor, the laser-modified region should be optically isotropic. It is well know that glasses can be poled under ultraviolet light or high dc bias condition at high temperature to create nonlinear optic behavior. Recent investigation on the local atomic structural change by an in-situ laser irradiation has demonstrated a polarization-dependent structure change in amorphous chalcogenide glasses. This field or polarization dependent mechanism is especially applicable to our fs laser processing since the estimated electric field generated by the laser pulse is on the order of 10¹¹ V/m. The strong electric field can effectively induce a strong contraction by an electrostrictive effect. Coupled with an immense localized heating and rapid quenching, this overall effect could result in an irreversible electrostrictive deformation and create an anisotropic densification that is analogous to the mechanism proposed for ultraviolet-induced densification of fused silica. It is believed that the anisotropic contraction of glass by the enormous field associated with fs laser pulses creates the optic birefringence in the bulk glass. Figure 8 shows a
microphotograph of a scanning electron microscopy (SEM) image on the photo-damaged region through fracture surface. Results show the photo-damaged regions have a periodic nano-scale structure consisting of glass and voids perpendicular to the beam direction. The laser generated nano-scale structure has a periodicity around 540 nm, which is independent of laser energy level and is very close to the wavelength of 548.7 nm as the 800nm fs laser pulses traveling in the silica. The creation of this periodic structure perpendicular to the beam propagation direction suggests an anisotropic contraction is induced in the field direction. The observation also confirms the Raman study where some voids have been created in the laser-modified regions. Because the creation of these voids in the laser modified region, the material near by the void will be put under pressure. The upward shift of Raman band for the 4-membered silica rings at 490 cm^{-1} seems to support the development of a compressive stress in the glass region. As a result, these glass regions in the periodic nano-scale structure might possess a higher density and a higher refractive index than the bulk glass. Furthermore, the periodic structure could also contribute to the development of optical birefringence. Birefringence of this nature is well known as ‘form’ birefringence. The Raman signatures for the photo-damaged and laser-modified waveguide regions in amorphous silica are extremely similar, if not identical. The slightly different can be attributed to a scalar factor that depends on the intensity of the laser pulse. Therefore, it is plausible that laser-induced anisotropic densification at the focus spot creates the optic birefringence and provides waveguide property in the bulk glass.

4. SUMMARY

Using a unique non-linear absorption behavior, we have demonstrated that when properly adjusted, femtosecond laser pulses can locally modify the glass structure and directly fabricate waveguides in bulk glass. Raman spectra indicate that femtosecond laser pulses create more three- and four-membered silica rings which are generally associated with quasai-surfaces at the small voids in the modified regions. Results show these laser-modified regions possess an optical birefringence property that can be controlled by the polarization direction of the laser beam. The phenomenon is explained in terms of an anisotropic densification by the immense electric field associated with the femtosecond laser pulse. Experimental observations from photo-damaged areas seem to support the explanation we have drawn. The capability of creating and controlling birefringence properties in glass by laser processing could have significant implications for the development of integrated novel optical devices.

ACKNOWLEDGMENTS

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Table I. The composition (weight percent), soft point, and glass transition temperature of different glasses

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<th>Glass Former</th>
<th>Glass Modifiers</th>
<th>Other oxides</th>
<th>T_s (°C)</th>
<th>T_g (°C)</th>
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<tr>
<td>SiO_2, B_2O_3, Al_2O_3</td>
<td>Na_2O, K_2O</td>
<td>MgO, CaO, BaO, ZnO, PbO</td>
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<td></td>
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<tr>
<td>Fused quartz</td>
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<td>0.00</td>
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<tr>
<td>CVD quartz</td>
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<td>0.00</td>
<td>0.00</td>
<td>0.00</td>
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<tr>
<td>Pyrex</td>
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<td>26.43</td>
<td>0.61</td>
<td>72.96</td>
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</table>
Figure 1. Micro-Raman spectra of laser-modified fused quartz.

Figure 2. Micro-Raman spectra of laser-modified on lead containing glasses.
Figure 3. Raman spectra from the Pyrex samples, unmodified substrate and femto-second-laser modified areas.

Figure 4. Direct-write buried waveguides in glass by femtosecond pulse laser. The top and bottom images are regions modified above and below the photo-damage threshold, respectively.
Figure 5 The amount of light scattering through the waveguide with different laser power (determined from top view image).

Figure 6. (a) The transmission image of laser modified regions under a cross-polarized light, the polarization direction of the laser beam is rotated 10° for each modification, (b) the change of transmitted light intensity in the first quarter of the circle.
Figure 7. The change of laser-induced birefringence in CVD quartz as a function of accumulated laser pulses.

Figure 8. SEM microphotograph of photo-damaged region on a three-point bend fracture surface (arrow – laser beam direction).
REFERENCES


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