



Higher refractive index and lower wavelength dispersion of SiO₂ glass by structural ordering evolution via densification at a higher temperature

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Large-sized densified silica glasses were fabricated at a high pressure of 7.7 GPa and high temperatures. As the synthesis temperature was increased from room temperature to 1200°C, the densification increased to 23%. The wavelength dispersion of the refractive index in visible region showed that not only the refractive index but also the Abbe number rose as high as those of crystalline silica as the synthesis temperature increased. The reflectance spectra in the vacuum ultraviolet region revealed that glasses densified at a higher temperature underwent homogeneous evolution of their structural ordering in an intermediate range. It was suggested that the decrease in randomness of the glass structure resulted in the increase in the refractive index and the Abbe number. The densification of glass at higher temperatures, which controls competing properties such as the refractive index and the Abbe number in the same direction, is a promising way to extract latent potential of materials.

Introduction

Silica (SiO₂) glass is an essential optical material. As it exhibits extremely low absorption and scattering losses during transmission of optical signals, optical silica glass fibers are used to transfer information across the Internet.^{1,2} Owing to its excellent optical transmittance properties in the UV region, silica glass is also used for lenses in semiconductor exposure apparatus, which govern the fabrication of advanced semiconductor chips with very thin wire electrodes.³

Silica glass is also a fundamental material in glass science. SiO₂ is a typical network-former oxide and can easily vitrify in a single component.⁴ The structure of silica glass has been investigated as the basis for amorphous materials and is generally viewed as a three-dimensional continuous random network of SiO₄ tetrahedra linked by corner-sharing oxygens,^{5,6} which resembles that of crystalline silica (α -quartz). However, as the network in silica glass is random, its density is lower (2.20 g/cm³) than that of α -quartz (2.65 g/cm³). Therefore, owing to its large free volume, silica glass can be substantially compressed under high pressure.⁷⁻¹⁰ An increase in density from 2.2 to 4.4 g/cm³ and an increase in the oxygen

coordination number of Si from four to six have been induced by applying pressures greater than 40 GPa.¹¹⁻¹³ After the pressure is released, the densification is relaxed, and the oxygen coordination number of Si returns to four. Nevertheless, the densification remains at ambient pressure, although the degree of densification is reduced. The maximum permanent densification of silica glass is approximately 21% after compression at 15–20 GPa at room temperature.¹⁴ The extent of the permanent densification depends not only on pressure but also on temperature.¹⁵⁻¹⁷ Greater compaction will be achieved with higher applied temperatures at a given pressure.

The densification of silica glass of up to 21% inevitably affects its optical properties. The refractive index determined using the Na D-line by the immersion method increased from 1.459 to 1.549 for 20% densification.^{18,19} However, the wavelength dispersion of the refractive index of highly densified silica glass has not been investigated, even though it is an important optical property for the accurate design of optics.^{20,21} This lack of data stems from the fact that the sample size used in high-pressure science is usually too small to allow the measurement of the sample's refractive index dispersion. Recent advances in high-pressure synthesis techniques have allowed us to use large-sized samples of silica glass for high-pressure and high-temperature experiments.²² In this study, large-sized densified silica glasses were obtained by using a high-pressure technique, and the optical properties of the densified glass were investigated.

Experimental procedures

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Densified silica glasses were obtained via high-pressure experiments by using a belt-type high-pressure apparatus.²³ The starting material was fused silica glass (T-4040, Covalent Materials Corp.). A glass sample with a diameter of 5.7 mm and a thickness of 1.5 mm was enclosed in a sample capsule that consisted of a tantalum sleeve, tantalum disks, and graphite lids. The glass was compacted under a pressure of 7.7 GPa, then heated at a given temperature for 30 min and cooled to room temperature over a period of 10 min. The pressure was relieved after the cooling period. The heating temperatures were room temperature, 400°C, 600°C, 800°C, 1000°C, 1200°C, and 1300°C.

The density of the glasses was measured using a gas pycnometer (AccuPycII 1340, Micromeritics). A precision refractometer (KPR-2000, Shimadzu) was used to measure the refractive index of glasses at various wavelengths. The vacuum ultraviolet reflectance spectra of the glasses and Z-cut α -quartz were obtained at the BL7B beamline of the UVSOR synchrotron radiation facility at the Institute for Molecular Science. All measurements were performed at room temperature.

Results and discussion

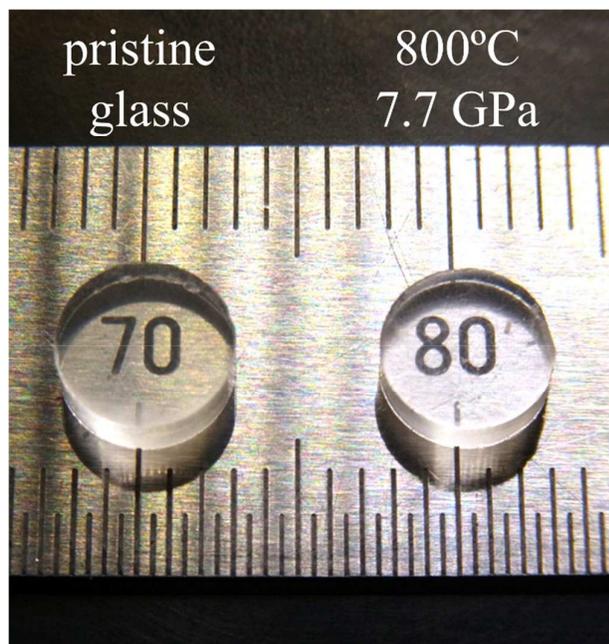
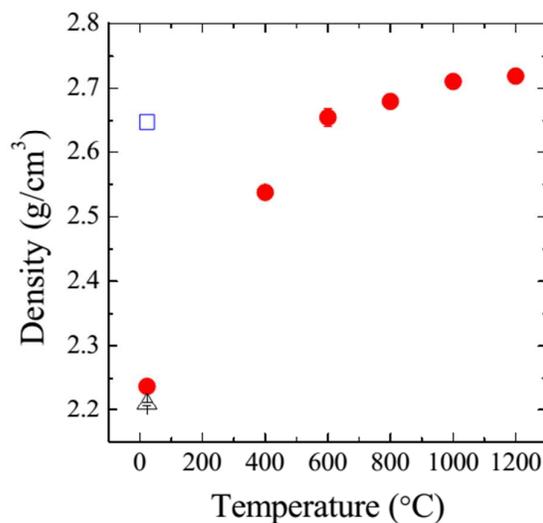


Fig. 1 (left) pristine glass and (right) glass densified at 800°C and 7.7 GPa.

The sample heated at 1300°C crystallized, whereas the other samples remained in the glassy state and were colorless and transparent. Figure 1 is a picture of pristine glass and glass densified at 800°C and 7.7 GPa. It is clearly seen that the densified glass is colorless and transparency comparable to the pristine glass, and the size of the densified glass shrank due to compression. Compared with the glassy state of samples reported in the previous study,¹⁶ the glassy state of our samples was stable at higher temperatures. The greater

stability of our samples may be due to their substantially larger size compared with the samples in previous experiments. The larger sizes of our samples may also have inhibited the crystallization. The sample preparation conditions and physical properties of the glasses prepared in the present study are summarized in Table 1.

Fig. 2 Densities of densified silica glasses prepared at various temperatures and 7.7



GPa. The open triangle and open square represent pristine glass and α -quartz, respectively.

Figure 2 shows the dependence of the sample density on the synthesis temperature. The density increased monotonically with increasing temperature. A densification of 20%, which resulted in silica glass with a density comparable to that of crystalline SiO_2 (α -quartz), was achieved even at a low temperature of 600°C. The increase in density apparently became saturated at temperatures greater than 600°C. The maximum density was 2.719 g/cm^3 , which is larger than that of α -quartz but does not reach that of coesite. The maximum densification of 23% is slightly greater than that reported previously.¹⁴ The larger maximum density might be the result of a combination of high pressure and high temperature. Further densification should be possible if a higher pressure can be applied to a large sample at a temperature of 1200°C.

Figure 3 shows the refractive indices of the samples at various wavelengths. Among a variety of polymorphs in crystalline SiO_2 , α -quartz is used here as a reference to the optical properties of densified silica glasses.²⁴ This is because densities of the densified silica glasses are around that of α -quartz and not close to other polymorphs. Additionally, it is rather difficult to obtain the precise data of optical properties such as wavelength dispersion of refractive index except for α -quartz because of the difficulty in making large-sized single crystals of other polymorphs such as coesite. The refractive index of the densified silica glasses clearly increases with the synthesis temperature. The refractive indices of the densified glasses synthesized at temperatures greater than 800°C are higher than those of α -quartz. The wavelength dispersion of

the refractive index of glass was approximated by using the three-term Sellmeier equation

$$n^2 - 1 = \sum_{i=1}^3 \frac{A_i \lambda^2}{\lambda^2 - \lambda_i^2} \quad (1)$$

where n is the refractive index at a given wavelength, λ (μm), and A_i and λ_i are fitting parameters. For silica glass, A_1 , A_2 , and A_3 are 0.6961663, 0.4079426, and 0.8974794, respectively and λ_1 , λ_2 , and λ_3 are 0.0684043, 0.1162414, and 9.896161, respectively.²⁵ The solid line of the calculated wavelength dispersion agrees well with the measured refractive indices of the pristine glass. The refractive indices at various wavelengths of the densified glasses were fitted by using the Sellmeier equation. The fitting was excellent, indicating that the refractive index dispersion of the densified silica glass was approximated well by the Sellmeier model.

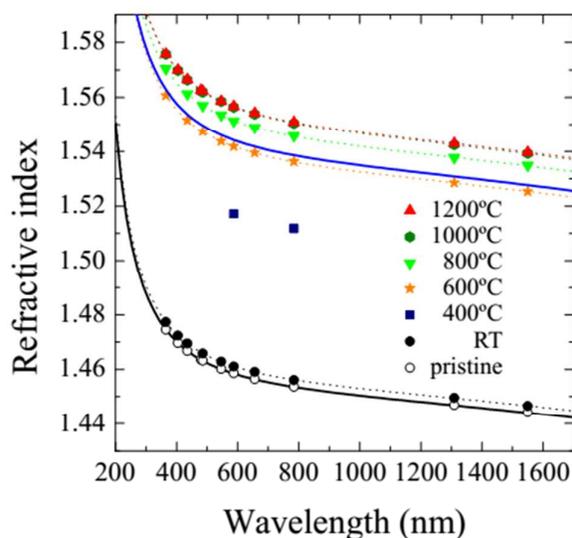


Fig. 3 Refractive index and wavelength dispersion of silica glass. Open circles represent pristine glass and solid symbols represent the densified glasses. The blue and black solid lines are the calculated refractive indices of α -quartz²⁴ and silica glass,²⁵ respectively.

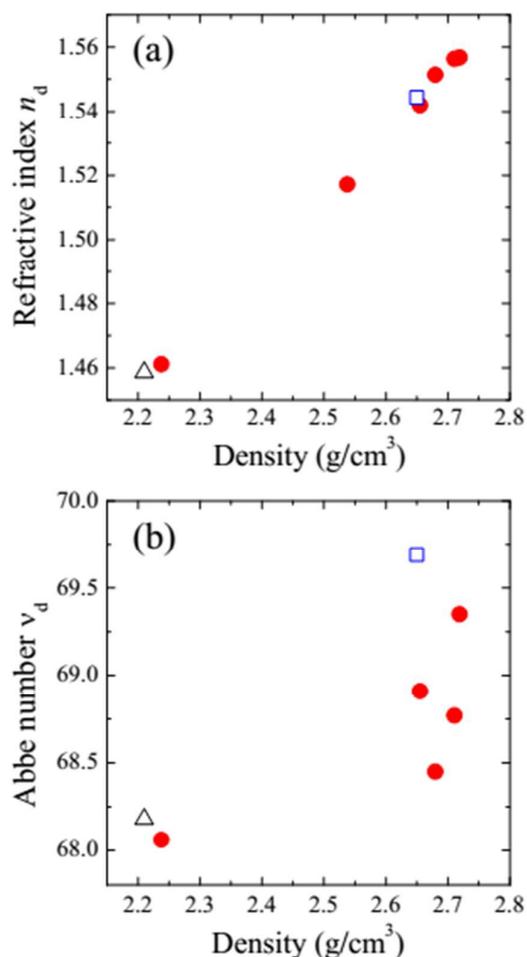
The refractive index at 587.56 nm, n_d , increased monotonically with the synthesis temperature and became saturated for samples synthesized at temperatures greater than 800°C, similar to the density. The maximum refractive index was 1.55680. The refractive index of a glass (n) is related to its density ρ according to the Lorentz–Lorenz equation^{26,27}

$$\frac{n^2 - 1}{n^2 + 2} \frac{M}{\rho} = \frac{4\pi\alpha_m N_A}{3} \quad (2)$$

where M is the molecular weight of the glass, α_m is the molar polarizability of the glass, and N_A is Avogadro's number. The density dependence of the refractive index is shown in Figure

4(a), where n_d linearly increases with increasing density. Although the density increases by 23%, n_d increases by only 6.7%. This difference is caused by the decrease in α_m from 6.8 to 6.2 Å³, which indicates that the Si–O bonding becomes more covalent.

The wavelength dispersion of the refractive index for optical glasses was evaluated on the basis of their Abbe number, $\nu_d = (n_d - 1)/(n_F - n_C)$, where n_F and n_C are the refractive indices at 486.1 and 656.3 nm, respectively. Lower wavelength dispersion results in greater values of ν_d . Although the data is slightly dispersed, ν_d increases monotonically with increasing temperature. The maximum ν_d is 69.4, which is smaller than that of α -quartz. These results indicate that if the densified glass exhibits a higher refractive index than α -quartz, the ν_d of the glass is always lower than that of α -quartz. The density dependence of ν_d is shown in Figure 4(b). The increase



in ν_d appears to occur primarily after densification. Fig. 4 (a) Refractive index at 587.56 nm (n_d) vs density. (b) Abbe number (ν_d) vs density. The open triangles and squares represent pristine glass and α -quartz, respectively.

The refractive index of α -quartz is plotted on the straight line in Figure 4(a), indicating that the refractive index of SiO_2 is dominated mainly by its density, irrespective of whether the SiO_2 is glass or crystalline. Therefore, a greater density leads to a higher refractive index. However, the wavelength dispersion of the refractive index of SiO_2 is governed by its absorption in the UV region. The main absorption is the exciton absorption observed at 10–11 eV.^{28–33} Figure 5 shows the reflectance spectra of the glasses in the vacuum ultraviolet (VUV) region together with the spectrum of Z-cut α -quartz. The intense broad peak at 10.1 eV is clearly seen in the spectrum of the pristine glass, whereas the α -quartz peak is narrower, more intense, and located on the high-energy side of 10.3 eV. This is because the electronic density of states of glasses is broader compared with that of the crystalline phase because of the amorphous random structure of glasses. The reflectance is related to the complex refractive index that is obtained by Kramers–Kronig analysis.^{31–33} Therefore, the lower wavelength dispersion of the refractive index of α -quartz compared with glasses is related to the reflectance change occurring more steeply at the high-energy side and the change is small in the visible region. The peak position of the reflectance might not have a significant effect on the refractive index dispersion because the peak is located far above the visible region and the difference in the peak position between glass and α -quartz is small. The steepness of the increase in the reflectance at around 10 eV increases in the order of glasses densified at 1200°C, 600°C, 800°C, 400°C, and the pristine glass, which corresponds to the order of the Abbe number. The steeper increase leads to the greater Abbe number and the lower wavelength dispersion of the refractive index. The experimental results from the reflectance spectra and the refractive index dispersion are consistent.

The main intense peak of the glass densified at 400°C is broader than that of pristine glass and shifted to a lower energy. This agrees well with the previous results for glasses that were 16% densified at 6 GPa and 500°C.³⁰ The lower energy side of the intense peak of the 16% densified glass was suggested to be a consequence of the decrease in the bandgap energy caused by the elongation of the Si–O bond length. The broadening of the peak of the 16% densified glass was probably due to an increase in the randomness compared with the undensified glass, because the intense peak of the undensified SiO_2 glass was broader than that of α -quartz. A molecular dynamics simulation of densified silica glasses showed a slight elongation in Si–O bond length, a significant decrease in Si–O–Si bond angles, and a decrease in the bond angle distribution of Si–O–Si after densification.³⁴ Accordingly, the increase in the randomness after densification, as observed in the reflectance spectra, was considered to be a result of the distortion of SiO_4 tetrahedra or the diversification of the rotational arrangements between SiO_4 tetrahedra.³⁰ These reasons can be applied to the case of the glass densified at 400°C in this study, because the experimental conditions and the densification value were similar to that of the 16% densified glass.

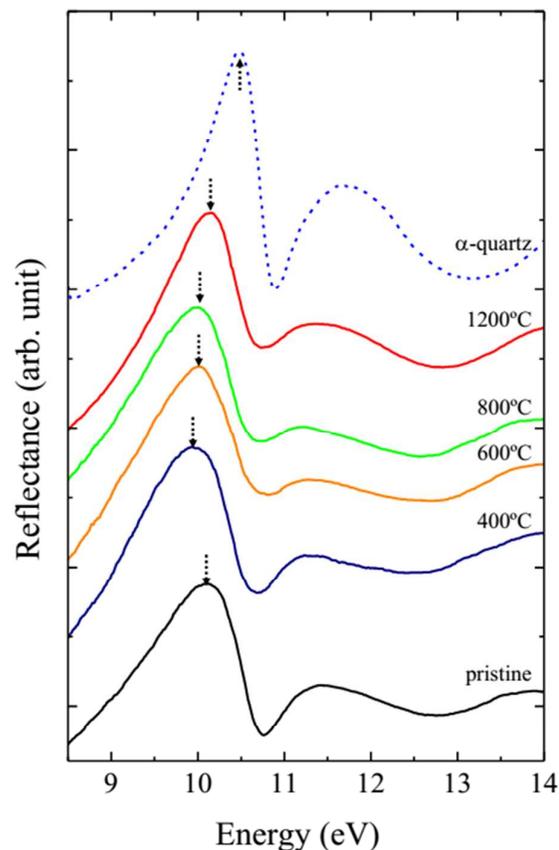


Fig. 5 Reflectance spectra of silica glasses and Z-cut α -quartz. The position of the intense peak is indicated by the dotted arrow.

The reflectance spectra of the densified glasses in this work show further interesting features. The intense peak shifted to the high-energy side and became sharper as the synthesis temperature increased. In particular, the glass densified at 1200°C had a sharper intense peak at a higher energy than the pristine glass. These observations contradict those in the previous report, indicating that the randomness of the glasses was decreased by densification at a higher temperature and the glasses became more homogeneous and ordered and close to a crystalline phase. Nevertheless, the glasses densified at higher temperatures are still amorphous, and the intense peaks of the glasses in the reflectance spectra are not sharper than that of α -quartz. This explains why the Abbe number of the densified glasses did not reach the value of α -quartz, even though it increased. These results indicate that the Abbe number of silica glass is dominated by the randomness of the structure. Accordingly, it may be possible to tune the refractive index dispersion of silica glass by adjusting the randomness of the atomic arrangement.

To investigate the change in structure from undensified glass to densified glasses, the structure factor $S(Q)$ obtained from X-ray diffraction measurements has been used. It was reported that there were obvious differences in the first sharp diffraction peak (FSDP) in the $S(Q)$, whereas there was no

difference in the high Q region.^{35,36} This means that the local structures such as the SiO_4 unit do not change after densification; however, the intermediate-range order structure is significantly different. In most cases, the densification experiments were performed at room temperature and the $S(Q)$ were obtained in *in situ* high-pressure experiments or from recovered glasses after densification. In those cases, the FSDP always broadened, the peak intensity decreased, and the peak position shifted to the higher Q side as the applied pressure was increased. Considering that the position of the FSDP is related to the structural correlation length for the intermediate-range order, the peak shift to the higher Q side indicates that the atoms became closer, or the atomic arrangements became more densely packed.^{14,35,36} The broadening of the FSDP and the intensity decrease resulted from the increase of the diversification of the intermediate-range order structure via densification.

On the other hand, in the $S(Q)$ obtained in *in situ* high-pressure and high-temperature experiments (9.9 GPa and 200–800°C), the sharpening of the peak and the increase of the peak intensity were observed, although the peak shifted to the higher Q side as in the case of the room temperature experiments. It was suggested that a more ordered, homogeneous structure at a higher temperature and a higher pressure was realized in the intermediate range.¹⁶ Heating a sample during the densification process has an important effect on the rearrangement of atoms to the less random structure because atoms can move more easily at a higher temperature, forming an orderly arrangement while retaining the amorphous structure. Accordingly, the randomness of the glass densified at 400°C becomes greater than that of the pristine glass, while the glass densified at 1200°C becomes more homogeneous and ordered than the pristine glass, which is consistent with the result from the VUV reflectance spectra. Considering that the density of the glass densified at 1200°C is larger than that of α -quartz, the atomic arrangement of the glass may be similar to that of coesite rather than α -quartz because coesite has a similar density of 2.9 g/cm³.^{37,38} Future structural analyses of the glasses densified at a higher temperature may find the coesite-like atomic arrangement characterized by four-membered rings. The quantitative structural analyses to quantify the randomness of the silica glass are now in progress.

Finally, to increase the refractive index of optical glasses in the visible region, some components with high polarizability and a high atomic weight, such as TiO_2 and La_2O_3 , were added to a base glass.^{39–41} These additives often narrow the optical bandgap, which results in the higher wavelength dispersion of the refractive index. Accordingly, a higher refractive index should lead to a higher wavelength dispersion. However, the wavelength dispersion of the densified silica glasses decreased even as the refractive index increased. The densification of glass at higher temperatures is a promising way to increase both n_d and ν_d , and this approach could lead to the fabrication of new advanced optical materials.

Conclusions

Large-sized densified silica glasses were fabricated under high temperatures and pressures. The maximum densification was 23% under synthesis conditions of 7.7 GPa at 1200°C. The large-size of the densified glasses allowed us to measure the refractive index with high accuracy enough to obtain the Abbe number ν_d . The refractive index of the glasses increased linearly with increasing density. The refractive index of the glasses was greater than that of α -quartz when their density was also greater than that of α -quartz. The Abbe number ν_d also increased with increasing densification, although it was always smaller than that of α -quartz. The reflectance spectra of the densified glasses and α -quartz in the VUV region showed that the randomness of the glass initially increased; however, it decreased as the synthesis temperature increased. This suggests that the densification at higher temperatures enabled silica glass to adopt a less random structure or it changed the structural ordering in the intermediate range. The increase in n_d and ν_d by densification was attributed to the decrease in the randomness accompanied by the densification at high temperatures. Consequently, it was proposed that even competing properties such as the refractive index and the Abbe number can be tuned by adjusting the randomness of the atomic arrangement. The densification at a higher temperature would lead to new findings in glass science.

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Table 1. Synthesis temperature, applied pressure, density, densification, refractive index at 587.56 nm (n_d) and Abbe number (v_d) of silica glasses. The data for α -quartz are also shown.

Temperature (°C)	Pressure (GPa)	Density (g/cm ³)	Densification (%)	n_d	v_d
RT	0 (pristine glass)	2.210	0	1.45859	68.2
RT	7.7	2.238	1.3	1.46109	68.1
400	7.7	2.538	14.8	1.51719	-
600	7.7	2.655	20.1	1.54194	68.9
800	7.7	2.680	21.3	1.55137	68.5
1000	7.7	2.711	22.6	1.55638	68.8
1200	7.7	2.719	23.0	1.55680	69.4
RT	0 (α -quartz)	2.648	-	1.5443	69.7

The silica glasses permanently densified at high temperatures show unexpected increase of both the refractive index and the Abbe number. It was suggested that glasses densified at a higher temperature underwent homogeneous evolution of their structural ordering in an intermediate range, which resulted in the unusual tendency of the optical parameters.

