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Pressure-induced structural changes in MgSiO$_3$ glass

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Introduction

It is necessary to know the structures of silicate melts when we understand the physicochemical properties such as density, viscosity, and elastic wave velocities. The structures of dense silicate glass, which is quenched from melt at high pressure and temperature or annealed below the crystallization temperature (T_c) at high pressure, have been investigated to predict the structures of silicate melts at high pressure (e.g., [1]; [2]). However, it is widely known that some of the structural changes in silicate glass are reversed during decompression. In-situ observation of structural changes under high-pressure conditions is essential for providing us with more precise knowledge of the structural evolution.

Experimental methods

MgSiO$_3$ glasses (*v-En*) were synthesized from MgSiO$_3$ enstatite powder by quenching the melt from 1923-1973 K. High-pressure and temperature X-ray diffraction experiments were performed at NE5C beamline with the MAX-80 DIA-type multianvil apparatus and tungsten carbide alloy anvils with 4- and 6-mm TEL (Truncated Edge Length). Temperatures were measured from EMF of the W3%Re-W25%Re thermocouple. The pressures were calculated form the EoS for NaCl. The diffraction data were collected over the Bragg angle of 3-30º to obtain wide range of scattering vector, Q ($Q=4\pi E\sin\theta/12.398[Å^{-1}]$, where E and q denote photon energy of X-ray and Bragg angle, respectively). The structure factors for *v-En* were obtained by using the analytical program developed by [3]. The radial distribution function, $G(r)$, was derived from Fourier transformation of structure factor.

Results and discussions

The T_cs at high pressure were determined by the appearance of diffraction peaks from MgSiO$_3$ phases (high and low clinoenstatite), in order to anneal the glass at high temperature. The T_c for *v-En* was found out to decrease with pressure, up to 9 GPa. This may indicate that the viscosity of MgSiO$_3$ melt decreases with increasing pressure to 9 GPa[4]. Since MgSiO$_3$ melt has relatively highly polymerized structure, this interpretation should be reasonable, as some previous studies have already shown.

Fig. 1 shows the pressure change of Si-O bond length. In the compressing at 300 K, the Si-O bond showed the minimum distance at 11 GPa. Then, the bond distance increased significantly up to 20 GPa. The bond length increase at pressures greater than 11 GPa indicates the coordination change of Si^{4+} to five and/or six. For instance, [5] predicted that 4 % of five-coordinated Si^{4+} appear in the *v-En* at 9 GPa and 300 K. However, such a structural change under room temperature condition has not been shown to be recoverable [2].

The Si-O bond length in annealed *v-En* began to increase at pressures greater than 2 GPa. This lengthening is related with the permanent structural change in *v-En* and can be interpreted as the change in chemical structure of silicate network. To test this interpretation, we measured Raman spectra of the recovered samples. In the sample recovered from pressure greater than 9 GPa, shoulder peak appeared on the low-frequency side of the main peak around 1000 cm^{-1}, which can be attributed to Q-speciation of SiO$_4$ tetrahedra. The shoulder peak was assigned more depolymerized chemical speciation of SiO$_4$[6]. In addition, such a pressure-induced depolymerization should result from the Si^{4+} coordination change. This observation suggests that the onset of Si coordination change is located between 2-9 GPa.
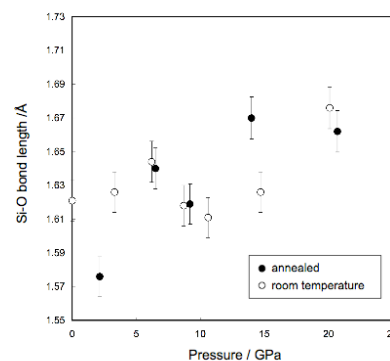

Fig. 1 Pressure dependence on the Si-O bond length

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