



# Contrasting sound velocity and intermediate-range structural order between polymerized and depolymerized silicate glasses under pressure



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## ABSTRACT

X-ray diffraction and ultrasonic velocity measurements of three silicate glasses (in jadeite, albite, and diopside compositions) show a sharp contrast in pressure-induced changes in structure and elasticity. With increasing pressure to around 6 GPa, polymerized glasses (jadeite and albite) display large shift in the first sharp diffraction peak (FSDP) in the structure factor,  $S(Q)$ , to higher- $Q$  values, indicating rapid shrinkage in the intermediate-range ordered (IRO) structure. Above 6 GPa, the shift of FSDP decelerates, suggesting that shrinkage in the IRO structure has been largely completed and the structure evolution is now dominated by the diminution of the interstitial volume in a more densely packed arrangement. Associated with this structural change, sound velocities increase with pressure above 6 GPa. In contrast, the depolymerized diopside glass exhibits smaller changes in the pressure dependence for both sound velocities and FSDP positions. Compared to the polymerized glasses, the velocities are faster and the positions of FSDP appear at higher- $Q$  under the same experimental conditions. The results suggest that the depolymerized diopside glass has an initially denser IRO structure compared to that of the polymerized glasses, and there are no sufficient interstitial voids to shrink. The different behaviors between polymerized and depolymerized glasses are apparently related to the initial linkage of tetrahedra and the pressure-induced structural reactions. These results suggest that under compression up to 10 GPa, the degree of polymerization is a major factor affecting the IRO network structure and the sound velocity of silicate glasses.

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## 1. Introduction

Igneous activities are primarily controlled by magmas, which occur in various geological/geophysical settings and have a wide range of compositions (e.g., basalt–andesite–rhyolite). These compositional differences largely determine structures of the magmas at depths and fundamentally control magma properties, which, in turn, control variations in eruption styles (e.g., Eichelberger et al., 1986). One of the most important parameters for constraining magma behaviors in the interior of the Earth is the degree of

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polymerization. Under upper mantle conditions, this is a useful indicator for the connectivity of SiO<sub>2</sub> tetrahedra, which are the basic units in all silicate melts. Compositional differences are generally considered in terms of variety of alkaline (e.g., Na<sup>+</sup>) and alkaline-earth (e.g., Mg<sup>2+</sup>, Ca<sup>2+</sup>) cations in the melts. These cations, called network modifiers, tend to cut Si–O–Si bonds between adjacent SiO<sub>4</sub> tetrahedra (e.g., Mysen et al., 1982). Al<sup>3+</sup> behaves as an intermediate species in silicate melts; it can act as either a network former (on the tetrahedral site) or a modifier, depending on composition (e.g., Riebling, 1966; Mysen et al., 1981). In the case of a fully-polymerized melt, such as jadeite and albite, all TO<sub>4</sub> tetrahedra are interconnected via oxygen anions, forming a continuous three-dimensional network. Whereas, for depolymerized diopside melt, approximately half the tetrahedral corners are disconnected. It is likely that the difference in the degree of polymerization results in different structural responses and physical properties of silicate melts.

Because of the technical difficulties in measuring structure and properties of silicate melts under high pressure and high temperature conditions, many researchers instead have made efforts for obtaining insights by studying glasses which are quenched from melts. The relationship between the degree of polymerization and the structure/properties of quenched (or supercooled) silicate melts under pressure has been reported previously (e.g., Allwardt et al., 2004, 2005, 2007; Lee, 2004, 2011; Lee et al., 2003, 2004, 2006, 2012; Xue et al., 1989, 1991). In addition to the quenched silicate glass studies at ambient condition, in-situ high-pressure structure and/or property measurements on silicate glasses have also been conducted. For example, silica glass ( $\text{SiO}_2$ ) has been well studied for pressure-induced changes in structure, density, and elastic properties (Susman et al., 1991; Meade et al., 1992; Zha et al., 1994; Sugai and Onodera, 1996; Tsiok et al., 1998; Clark et al., 2004; Inamura et al., 2004; Sato and Funamori, 2008, 2010; Murakami and Bass, 2010; Yokoyama et al., 2010; Kono et al., 2011). Sound velocities of  $\text{SiO}_2$  glass up to 57.5 GPa have been investigated using the Brillouin scattering measurement (Zha et al., 1994). In this latter study, a negative pressure dependence in velocities up to about 3 GPa is reported, followed by a drastic increase in the velocities between 12 and 23 GPa. Ultrasonic velocity measurements have also been performed on  $\text{SiO}_2$  glass (Yokoyama et al., 2010; Kono et al., 2011). In both studies, anomalous minima in  $V_P$  and  $V_S$  around 3 GPa were also found, confirming the Brillouin scattering measurements.

However, the effect of degree of polymerization in silicate melts/glasses on the structural response and physical properties under high pressure remains unclear (e.g., effect of degree of polymerization on the IRO, sound velocity, density, as well as pressure dependence of these properties). For silicate melts at ambient pressure, the degree of polymerization is closely related to composition and is quantitatively described by a ratio of non-bridging oxygens (NBO) to tetrahedral cations (T) (NBO/T; Mysen et al., 1982). The NBO/T ratio is widely used in examining viscosity systematics in various silicate melts and discussing magma mobility in the Earth's interior (e.g., Suzuki et al., 2005). Experimental data show that polymerized melts generally possess much higher values of viscosity than those of depolymerized ones near 1 atm (Dingwell and Virgo, 1988; Richet et al., 1996; Dingwell et al., 2004) and that pressure dependences of the viscosities for polymerized and depolymerized melts are fundamentally different: while viscosities of polymerized melts decrease with increasing pressure, those of depolymerized melts increase (Taniguchi, 1992; Wolf and McMillan, 1995; Poe et al., 1997; Funakoshi et al., 2002; Reid et al., 2003; Liebske et al., 2005; Suzuki et al., 2005, 2011; Lesher, 2010; Sakamaki et al., 2013). Although NBO fraction in the silicate glass generally decreases gradually with pressure (e.g., Lee, 2011), NBO/T under ambient condition appears to be a useful indicator for understanding the behavior of silicate melts.

In this study, we investigate sound velocities of polymerized (jadeite and albite composition: NBO/T = 0 at 1 atm) and depolymerized (diopside composition: NBO/T = 2 at 1 atm) glasses at pressures up to 10 GPa by using ultrasonic technique and synchrotron X-ray imaging in a Paris–Edinburgh press. We also study X-ray structure factors,  $S(Q)$ , of these glasses by using the multi-angle energy-dispersive X-ray diffraction technique, in order to understand structural changes in the intermediate-range order (IRO) with pressure. Finally, we discuss the correlation between sound velocities and structures, and the influence of the degree of polymerization on these properties.

## 2. Experimental methods

All experiments were conducted in a Paris–Edinburgh press, installed at the HPCAT 16-BM-B beamline, Advanced Photon Source

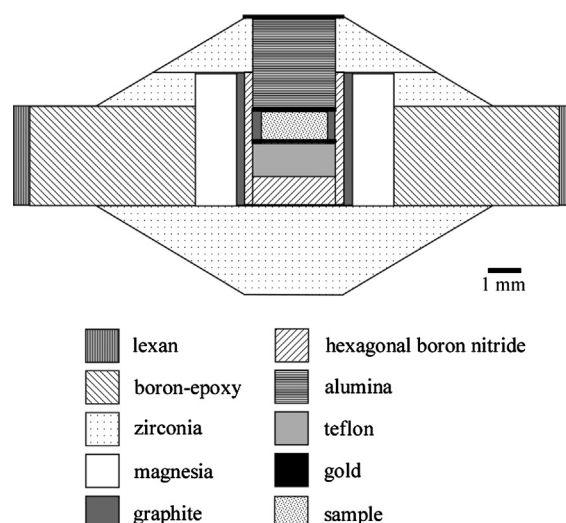


Fig. 1. A design of cell assembly for ultrasonic measurements.

(APS). Details of the setup are given by Kono et al. (2013). A schematic illustration of the high-pressure cell assembly is shown in Fig. 1. The outer pressure media consisted of machinable zirconia pallets and sintered boron-epoxy. Graphite cylinder was used as a sample container, with a gold foil placed on top and bottom of the capsule as markers for sample length measurements.

X-ray radiography images allowed us to determine sample lengths under high pressure (Fig. 2a). A fully densified alumina cylinder above the sample was used as a buffer rod for the transmission of ultrasonic signals. Both the glass sample and the alumina buffer rod were polished with 1  $\mu\text{m}$  diamond paste to ensure good mechanical contact. High-pressure sound velocity measurements were carried out using the ultrasonic pulse-echo-overlap method. Details of the ultrasonic measurement are described in Kono et al. (2012). Ultrasonic signals were generated and received by a  $\text{LiNbO}_3$  transducer. The signals were collected with a sampling rate of  $5 \times 10^9$  Hz (Fig. 2b). The compressional and shear wave travel times were determined using the reflected signals from the buffer rod/sample (R1) and sample/teflon (R2) interfaces. The travel time was determined within the uncertainty of  $\pm 0.1$  ns, corresponding to an uncertainty in compressional and shear wave velocities of up to  $\pm 0.08\%$  and  $\pm 0.05\%$ , respectively. Uncertainty of the sample length determination was less than  $\pm 0.948$   $\mu\text{m}$  ( $\pm 1$  pixel) in a conservative estimation, because the buffer rod/sample and sample/backing reflector interface position was determined with the standard deviation of less than  $\pm 0.190$   $\mu\text{m}$  ( $\pm 0.2$  pixel), respectively. The  $\pm 0.948$   $\mu\text{m}$  uncertainty in sample length determination corresponds to  $\pm 0.26\%$  error in both compressional- and shear-wave velocity determinations. We therefore consider the overall uncertainty in the compressional and shear wave velocity measurement to be less than  $\pm 0.34\%$  and  $\pm 0.30\%$ , respectively.

Structure measurements were performed using the energy dispersive X-ray diffraction (XRD) technique as described in Yamada et al. (2011). The 16-BM-B is a bending magnet beamline providing white X-rays (5–120 keV) with high brightness. The incident X-ray beam was collimated by two sets of vertical (0.1 mm) and horizontal (0.1 mm) slits. The diffracted signal was collimated with a 0.1 mm gap collimator, 60 mm downstream from the sample, and a 0.1 mm (H)  $\times$  5.0 mm (V) receiving slit 480 mm further downstream from the scattering slits. A Ge-SSD with a 4096 multi-channel analyzer was mounted on a two-theta arm, on air pads over a granite table, on a large Huber rotation stage, which allowed accurate control of the two-theta angles. Diffraction patterns were collected at 9 fixed diffraction angles ( $2\theta = 3^\circ, 4^\circ, 5^\circ, 7^\circ, 9^\circ, 11^\circ, 15^\circ, 20^\circ, 25^\circ$ ). Collecting time varied with

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