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高圧下における珪酸塩ガラスの音速・構造測定
Sound velocity and structure measurement of silicate glasses under pressure

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The degree of polymerization in silicate melt/glass is one of the most important parameters to understand the magma behavior. For silicate melts at ambient pressure, the degree of polymerization is highly related to composition, which is quantitatively described by a ratio of non-bridging oxygen (NBO) to tetrahedrally cation (T). In particular, the NBO/T is widely used to obtain viscosity information of various silicate melts and discuss the magma mobility in the Earth's interior. Several viscometry studies reported that polymerized melts showed much higher values of viscosity than those of depolymerized ones. Interestingly, it should be noted that the pressure dependence of the high viscosity of polymerized melts was shown to be negative. This gives important questions of the compression effect on the degree of polymerization and its effects on properties of silicate melts. In this study, we have measured the sound velocity of polymerized glass (jadeite and albite glass: NBO/T=0) and depolymerized glass (diopside glass: NBO/T=2) at pressures up to 10 GPa by using ultrasonic technique and synchrotron radiation with a Paris-Edinburgh press. We have also obtained the X-ray structure factor, S(Q), of these glasses by using energy-dispersive X-ray diffraction method in order to understand structural changes in the intermediate-range order with pressure.

All experiments were conducted using a Paris-Edinburgh press, which is installed at the HPCAT 16-BM-B beamline, Advanced Photon Source (APS). High pressure sound velocity measurements were carried out using the ultrasonic pulse-echo-overlap method. 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 measurement. Radiography images taken by CCD camera allowed us to calculate the sample length under high pressure. Pressure was determined by the equation of state of gold, which was located below the sample. The scattered X-rays were detected using a Ge solid state detector (Ge-SSD) with a 4096 multi-channel analyzer. Alumina above the sample was used as a buffer rod. The glass sample and the alumina buffer rod were polished with 0.001 mm diamond paste. Ultrasonic signals were generated and received by a LiNbO3 transducer. The signals were collected with a sampling rate of 5 x 109 point/second. Structure measurements were performed using the energy dispersive X-ray diffraction technique. The 16-BM-B is a bending magnet beamline which provides white X-rays (5-120 keV) with high brightness. The incident X-ray 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 scattering slit 80 mm downstream from the sample and a 0.1 mm x 5.0 mm receiving slit 400 mm further downstream from the scattering slit. The Ge-SSD was mounted on a two-theta arm on a large Huber rotation stage, which allows accurate control on two-theta angle. The diffraction patterns were collected for 9 fixed diffraction angles (2theta = 3, 4, 5, 7, 9, 11, 15, 20, 25 degrees). Collecting time varied with the diffraction angles, as intensities decreased with increasing angle. All patterns were collected until the maximum intensity reached at least 2000 counts. Structure factor, S(Q), was obtained by combining X-ray diffraction profiles collected for 9 diffraction angles.

Pressure dependence of sound velocity of jadeite, albite and diopside glasses will be presented, along with structure factor S(Q) of the glasses at high pressure. We would like to discuss a direct correlation between the intermediate-range order structure and sound velocity in these glasses, and the influence of the degree of polymerization.

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