

SCG009-P01

Room:Convention Hall

Time:May 23 10:30-13:00

Application of the EXEFS to the structure of the high-pressure aluminosilicate glasses

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Silicate melts play an important role in the chemical evolution of the planetary mantle during the early magma ocean stage as well as the subsequent long history. Movement of silicate melt in the mantle is controlled by its viscosity and density that are related to the melt structure. Therefore, the structural study of silicate melts under pressure is fundamental to understand the magma-related phenomena within the planets. Pressure-induced structure change in silicate melts have been studied with the quenched glass by using spectroscopic methods such as the NMR and the XAFS. Here we report the first results of application of the extended x-ray emission fine structure (EXEFS) to the structure analysis of the quenched silicate glass. The EXEFS arises from the radiative Auger effect and has the same structure as the XANES. The EXEFS can be measured using a wavelength-dispersive electron microprobe.

We measured the EXEFS spectra of the $\text{Ca}_3\text{Al}_2\text{Si}_6\text{O}_{18}$ (CAS) composition glasses quenched at 0.1MPa and 8 GPa. High-pressure glass was prepared by using a KAWAI type multi-anvil apparatus. The Si EXEFS spectra show that the silicon in the CAS glasses takes four fold coordination up to 8 GPa. On the other hand, the coordination change of aluminum from four to five is detected by the EXEFS, which is consistent with the results of the Al NMR study on the same composition quenched glasses (Allward et al. 2005).

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Density measurement of liquid Fe-Si using X-ray absorption method

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The density of liquid Fe alloys under high pressure is important for estimating the amount of light elements in the Earth's outer core. Here, we performed the density measurement on the liquid Fe-10wt%Si using X-ray absorption method in order to clarify the effect of pressure on the density and to determine the equation of state of the liquid.

X-ray absorption method provided the density of the liquid Fe-10wt%Si at pressures and temperatures up to 3.6 GPa and 2173 K, respectively. The density of the solid Fe-10wt%Si decreased with increasing temperature (1073-1373 K). However, the density of liquid Fe-10wt%Si did not show a clear tendency to the temperature. The thermal derivative of the density of this study was $-0.00055 \text{ gcm}^{-3}\text{K}^{-1}$ at 3.5 GPa, whereas that of ambient pressure was $-0.001 \text{ gcm}^{-3}\text{K}^{-1}$. Therefore, the effect of temperature on the density of the liquid under high pressure is much weaker than that of the ambient pressure. Vinet equation of state yielded isothermal bulk modulus $K_{0T} = 59(5) \text{ GPa}$ with its pressure derivative $K' = 4$ at 1873 K. The present results revealed that the substitution of Si into Fe decreases not only the density of liquid Fe but also the bulk modulus of that. Based on the obtained density and bulk modulus of liquid Fe-Si, the bulk sound velocity (V_P) of liquid Fe-Si is lower than that of pure liquid Fe in the range of our experimental condition.

Keywords: high pressure, density, light element, X-ray absorption method, equation of state

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Structure of jadeite-diopside melts at high pressure by in situ x-ray diffraction

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Properties of silicate melts control magma-related processes such as volcanic activity and evolution of the Earth. Since these processes take place in Earth's deep interior, there is considerable interest in documenting experimentally how pressure affects properties of silicate melts. Macroscopic physical properties are largely determined by the microscopic structure. The bond length and strength between tetrahedrally coordinated cation (T=Si⁴⁺, Al³⁺) and oxygen (T-O length) are especially important in the relationship between structure and physical properties of silicate melts. For silicates, T-O lengths are the shortest among a large variety of melts, therefore we need XRD data with large Q coverage in order to obtain accurate T-O length experimentally. In this study, we tried out XRD experiments by Paris-Edinburgh press, which enables us to get XRD patterns to 2theta angles as high as 40 degree, and photon energies in excess of 100 keV. On the basis of the structural investigation at ambient pressure, the jadeite melt is a typical polymerized melt, while the diopside melt is depolymerized. Considering the structural parameters under ambient condition the ratio between non-bridging and tetrahedrally bonded oxygen (NBO/T), jadeite melt is 0 and diopside melt is 2. Therefore, these two compositions would allow us to examine the relationship between structure and composition of silicate melts.

High-pressure and high-temperature XRD experiments were carried out in the Paris-Edinburgh press, which was developed by GSECARS and installed at the HPCAT beamline 16-BM-B of APS. The compositions of starting materials were synthetic jadeite (Jd), diopside (Di), and Jd₅₀Di₅₀. The sample container was graphite. The encapsulated samples were enclosed in an hBN cylinder, which served both as an electric insulator and a pressure marker. High-temperature was generated by resistive heating of graphite heater outside the BN cylinder. Pressure medium consisted of ZrO₂, MgO and boron-epoxy. The center of the pressure medium was boron-epoxy and MgO, because of their low absorption to X-ray. The incident X-ray was collimated by a vertical slit (0.5 mm) and a horizontal slit (0.1 mm) to irradiate the sample. The diffracted X-ray was detected by a Ge solid state detector with a 4000 multi-channel analyzer, through vertical (0.5 mm) and horizontal (0.1 mm) receiving slits as well as a collimator. The diffraction patterns were collected for 12 fixed diffraction angles (2theta=3, 4, 5, 7, 9, 11, 15, 20, 25, 30, 35, 39.5 degrees).

The structure measurements of jadeite-diopside melt were carried out in the pressure range from 1 to 5 GPa and at 1600 to 2000 K. Results on structure factors S(Q) and radial distribution functions G(r) of these melts at high pressures and high temperatures will be discussed.

Keywords: melt, high pressure, high temperature, structure, X-ray diffraction

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Molecular dynamics simulations of sodium silicate melts

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Inter-atomic potentials are essential for precise reproduction and prediction of physical property of the system using molecular dynamics simulation. In the silicate system, many pair potential models have been proposed. Some of these can reproduce crystal structures and its elastic properties (e.g. van Beest et al. 1990) and were used for silicate melts (Lacks et al. 2005). However, these previous models which can reproduce qualitative pressure dependence of viscosity seem not to reproduce temperature dependence of Q-species (Maehara et al. 2005). Understanding the physical properties and their characteristic behavior of silicate melts, nano-structure and its temperature, pressure and composition dependence are needed to know.

To investigate the nano-structure of silicate melts, we employed newly developed inter-atomic potential model for silicate systems. Our potential model definitely includes coulombic interaction, short range repulsion, van der Waals interaction and radial covalent interaction terms. Molecular dynamics simulations were performed for $\text{Na}_2\text{Si}_2\text{O}_5$ system using the MXDORTO. The number of atoms, pressure and temperature are maintained constant (NPT ensemble, $N_{\text{atom}}=5994$, $P=0.1\text{MPa}$). Physical property and Q-species were obtained at every 300K during the cooling from 3000K. The equilibrium data were obtained after 2-3ns (2-3,000,000steps) relaxation at each temperature. From this simulation, thermal expansion and temperature dependence of Q-species were investigated. The temperature dependence of Q-species was qualitatively reproduced, however it was not enough yet at quantitative aspect.

To improve the inter-atomic potential parameters, we are performing MO calculation using Gaussian09. To focus on Si-O-Si bond, we calculated energy surfaces of silicate clusters contain Si-O-Si bond (e.g. dimer, rings) for structural changes. By fitting inter-atomic potential parameters to these energy surfaces, improvement of reproducibility of physical properties is expected and the relation between physical properties and atomic structure might be discussed.

Keywords: MD, molecular dynamics, silicate melt