Structural investigation of $NaAlSi_3O_8$ melt under high pressure and high temperature

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1. Introduction

Feldspar group is one of the most major minerals in the Earth's continental crust. Albite ($NaAlSi_3O_8$), which is the end member of Na_2O, is thought to be the key of the magma genesis because soda lowers the melting temperature significantly. The physical properties (e.g., density and viscosity) of albite melt have been measured with several techniques of in situ and/or ex situ. The results of viscometry have shown an anomalous behavior at lower pressure region. In specific, the viscosity decreases with increasing pressure at least up to ~3 GPa (e.g., Kushiro et al., 1978; Suzuki et al., 2002), then eventually, it may turn to increase at higher-pressure region. The decrease in viscosity of albite melt at lower pressure has been explained with the decrease in the bond angles of alminosilicate network and/or coordination number change in aluminum ion (i.e., four to five or six coordinated Al^{3+}). The structural change of aluminum in the melt has been proposed at around 3 GPa, from the evidence that albite transforms to jadeite at 3 GPa (Waff, 1979), which has six-coordinated aluminum site, and that the melt viscosity decreases at low-pressure region as described above (Kushiro et al., 1978). However, structural investigation by Raman spectroscopy (Sharma et al., 1989; Mysen et al., 1983) on the glass recovered from 3 GPa and superliquidus denoted no evidence for the existence of highly coordinated aluminum. On the other hand, similarly, the structure of glass recovered from higher pressure, ~8 GPa, has been reported to include very small amount of highly coordinated aluminum such as 5- and 6-coordinated one. Here, these previous structural studies have been performed on recovered glass sample, which means that the recoverability of glass/melt structure is not taken into account. In order to investigate precisely the pressure where aluminum coordination transformation happens, the structural change should be monitored in situ at high pressure and temperature.

We have conducted in-situ structural analysis on hydrous and anhydrous albite melt by high-pressure and temperature x-ray diffraction with synchrotron radiation plus DIA-type large volume press (LVP) at PF-AR. The radial distribution analysis has indicated slight increase in T-O bond length in anhydrous melt (T means Si and Al atoms) at around 3 GPa, suggesting that the coordination number of Al^{3+} increases. On the other hand, clear feature that could be attributed to highly coordinated Al was observed in hydrous melt, specifically the shoulder peak appeared on the long-distance side of first peak in radial distribution function (~1.7 Å). In addition, the shoulder peak becomes more intense with increase pressure. From our in-situ observations, it is possible that coordination change in aluminum ion in melt is occurred at much lower pressure than one as expected ex-situ glass structure study. However, unfortunately, it is difficult to conclude that aluminum coordination change happens at around 3 GPa only from XRD, especially for anhydrous melt because of resolution of radial distribution function, RDF, (Si-O and Al-O peaks cannot be distinguished). To solve this issue, neutron diffraction is quite promising technique because diffraction data can be obtained over wide scattering vector (>30 $Å^{-1}$), which means that the resolution of RDF can be dramatically improved. Additionally, the diffraction data by neutron give us more specific information about Al by compensating the data set from XRD due to the difference of scattering length.

2. Experiment

High-pressure and temperature experiments were performed with 6-rams LVP (Large Volume Press), ATSUHIME. The 6-6-type cell with 10.0-mm TEL anvils was adopted for the present experiment. The graphite heater and the sample capsule shown in Figure 1 are enclosed in 17-mm cubic pressure transmitting medium made

of zirconia. The temperature at high pressure has been estimated from power consumption vs. temperature relationship by preliminary experiment using the same cell assembly with W3%Re-W25%Re thermocouple.

High-pressure diffractometry for albite melt has been conducted at 3.0 and 1.6 GPa (420 and 800 kN /ram), which corresponds to ~3 and ~5 GPa, and just above their melting temperatures, < ~1600°C. the pressures were calculated from equation of state of MgO at room temperature. The melting of the sample was confirmed by the diffraction pattern with elevating temperature. In specific, disappearance of sharp diffraction peaks from the crystalline sample was monitored with elevating temperature. In order to estimate initial source intensity, I_0, vanadium put in the same assembly was measured at high pressure and temperature. The empty cells were also measured for the correction on background at 1 atm. The deformation of the cubic cell and the fins at high pressures were estimated from the stroke of rams. Diffracted neutron was detected by position sensitive detectors (PSD) located at 90 ±11.3 degree through the gap of the second anvils. Almost only diffraction from sample can be collected due to good collimation with the radial collimators. The exposure time for the diffractometry for the melt was up to ~30 hours.

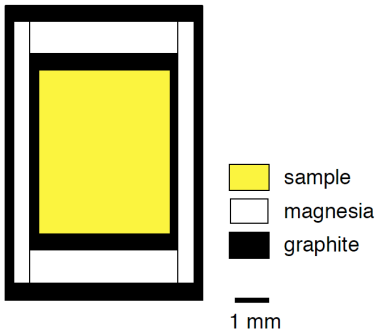

Figure 1. The cross section of sample room in zirconia pressure transmitting medium

3. Results

Structure factor for albite melt at ~5 GPa was plotted with the one taken at ~1 GPa (from last beamtime) in Fig. 2. The first sharp diffraction peak (FSDP, ~1.5 $Å^{-1}$) in ~5 GPa clearly shifted toward higher-Q side and showed weakening. The weakening of FSDP is widely observed in densified network-forming glasses such as GeO_2 and SiO_2. Radial distribution functions, g(r), which can be derived from Fourier transformation of S(Q), are listed in Figure 3. Although the g(r)s still include small noise, first intense peak attributed to T-O atomic pair (T= Si and Al) exhibits broadening at high pressure, suggesting that Al^{3+} partially transforms to higher coordination states.

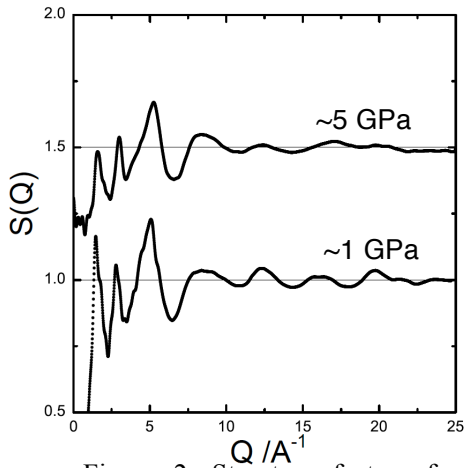

Figure 2. Structure factors for albite melt at high pressure.

4. Conclusion

In the present beamtime, overall, we have successfully collected diffraction of albite melt up to ~5 GPa. The local structure of albite melt with pressure would be clarified combining with XRD data although we need to check the diffraction data collected in this beamtime carefully. Regarding the future work, high-pressure neutron diffraction study would be next target.

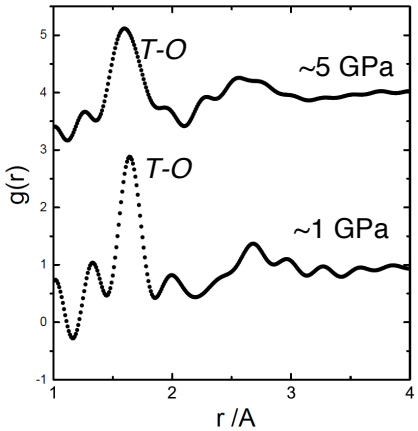

Figure 3. Radial distribution function for albite melt at high pressure.