CROSS TOKAI Experimental Report J-PARC	承認日 Date of Approval 2017/1/17 承認者 Approver Takanori Hattori 提出日 Date of Report 2017/1/17
課題番号 Project No. 2015A0181 実験課題名 Title of experiment Structural investigation of $NaAlSi_3O_8$ melt under high pressure and high temperature 実験責任者名 Name of principal investigator Akihiro Yamada 所属 Affiliation Center of Glass Sci. Technol., The Univ. of Shiga Pref.	装置責任者 Name of Instrument scientist Takanori Hattori 装置名 Name of Instrument/（BL No.） BL11 実施日 Date of Experiment 2016/12/9-12/16

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Sample for the present experiment has been prepared from Na_2CO_3, Al_2O_3 and SiO_2 to be $NaAlSi_3O_8$ composition, which corresponds to the major component of the Earth's crust (albite). The oxide mixture was decarbonized at 900°C for 9 hours. Then, the sample was sintered with the shape of column (ϕ4.6 mm) at 650°C for 10 hours, so as to be shrunk to ~4-mm diameter. Finally, the sintered sample was loaded in the graphite cup (ID: 4.0, H: 5.0 mm).

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

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 (410 and 210 kN /ram) and just above their melting temperatures, < ~1450°C. The melting of the sample was confirmed by the diffraction pattern. 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. Neutron diffraction were taken by position sensitive detectors (PSD) located at 90 degree ± 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 about 10 hours, due to the heater trouble. In specific, the resistivity of heater got suddenly high and finally current was cut to almost zero. It is likely due to the thinning of the graphite heater because of the diffusion of graphite into the zirconia, which surrounds the graphite heater.

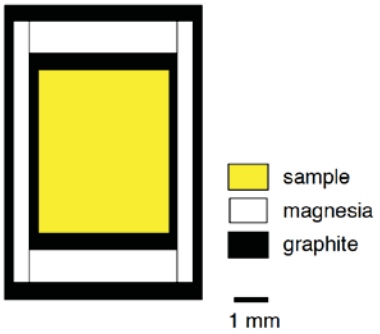

Figure 1. The cross section of sample room in zirconia pressure transmitting medium

The structure factor, S(Q), of albite melt taken at ~1 GPa is shown in Fig. 2. The diffraction of albite melt at high pressure could be successfully collected with analyzable signal/noise ratio up to ~25 $Å^{-1}$. The radial distribution function, g(r), can be derived from the Fourier transformation of S(Q), as shown in Fig. 3. The striking peak at around 1.6 Å is from T-O nearest neighbor in albite melt (T= Si, Al). At 2.6 Å, O-O pair can be clearly observed, of which intensity is more pronounced in neutron diffraction than that of x-ray diffraction. On the other hand, the intensity of peak at around 3.1 Å, which is from of T-T pair, is much weaker than that of x-ray. This observation is consistent previous studies on neutron diffraction experiment for silicate glass. The bond length of T-T obtainable from the present g(r) is 1.644 Å, which is slightly longer than that of Si-O in silicate (Al-free) glass and/or melt. This result could indicate that the interatomic distance of Al-O in the albite melt is longer than Si-O. The Al-O distance could be lengthened at higher pressure due to the Al coordination change, such as four-fold one to five- or six-coordinated Al. It may be expected that the T-O peak can be split as doublet when the Al coordination number becomes higher due to the longer interatomic distance in AlO_n polyhedra such as AlO_5 and/or AlO_6.

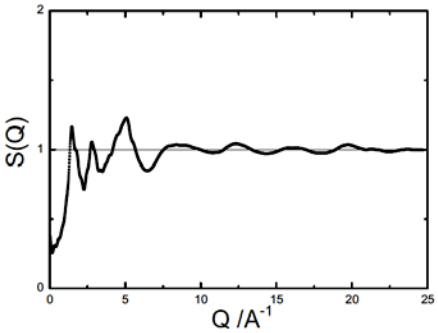

Figure 2. The structure function, S(Q), of albite melt at ~1 GPa.

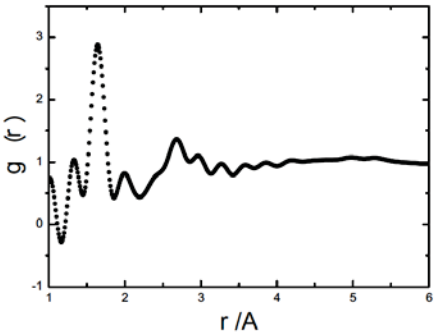

Figure 3. The radial distribution function, g(r), of albite melt at ~1 GPa.

In this beamtime, we have successfully demonstrated the usability of the combination of ATSUHIME with the intense neutron source by J-PARC for high-pressure structural analyses on silicate melt, which includes a lot of experimental difficulties. In order to achieve the direct observation at higher pressure (e.g., Al coordination change), intensification of the neutron source at J-PARC is strongly desired.