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In situ X-ray experiment on hydrous Mg silicate melts under high-P-T conditions using single crystalline diamond capsule

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Introduction

Recent studies on melting of mantle minerals have revealed that the liquidus phases change from olivine to pyroxene, and further stishovite with increasing pressure under hydrous condition, and the compositions of the liquid generated in the deep mantle become enriched in MgO component (e.g. Inoue, 1994[1], Yamada et al., 2004[2]). This phenomenon implies the structural changes of the hydrous silicate melts in the vicinity of the pressure at which liquidus phases are changed, because the liquidus phases and the melting relations should be strongly affected by the melt structures. In spite of the importance, however, direct observation on hydrous silicate melts under high pressure has not been conducted yet because of the experimental difficulties. A few studies were conducted so far by analyzing hydrous silicate glasses quenched from hydrous melt at high pressure (e.g. Xue et al., 2004[3]). We have conducted in-situ X-ray diffraction experiments of hydrous Mg-silicate melts under high pressure and high temperature to constrain the structures. Special attentions were made to obtain the diffraction by introducing the new diamond capsule system.

Experimental

High pressure and high temperature experiments were conducted at AR-NE5C bending magnet beamline, using MAX-80 cubic type high pressure apparatus. The anvil truncation edge length was 6 mm. X-ray diffractions of molten hydrous silicates were measured by white X-ray (5-140 keV) energy dispersive method using Ge-SSD. Diffraction angles were fixed at several arbitrary angles between 3° and 25° to obtain the data in wide scattering vector Q range (Q (Å^{-1}) = $4\pi E\sin\theta/12.398$, where E is photon energy).

Starting materials were prepared by mixing of $Mg(OH)_2$, MgO and SiO_2, to make the systems of Mg_2SiO_4-H_2O (Mg/Si=2.0, H_2O=20.4 and 11.3 wt%), Mg_2SiO_4-$MgSiO_3$-H_2O (Mg/Si=1.5 H_2O=18.3 wt%) and $MgSiO_3$-H_2O (Mg/Si=1.0, H_2O=15.2 wt%). Pressure transmitting medium were made of Boron + epoxy resin (4: 1) or pyrophyllite with the shape of 9 mm cube. Prior to X-ray diffraction experiments, we carried out temperature calibration experiment using the same cell assembly with W3%Re-W25%Re thermocouple. Hydrous molten samples were enclosed by single crystalline diamond sleeve (cut in parallel to (100)) combining with rare-metallic lids made of platinum. Both ends of diamond sleeve were sealed by platinum cap. Pressures were calculated by Pt and MgO markers put on the top of sample chamber.

Results

We could succeed to get good quality diffraction data of hydrous silicate melts up to 6 GPa by using diamond capsule, and also could 8 times reduce the exposure time comparing with the use of Ag-Pd capsule, in which we have done experiments previously.

Structure factor S(Q) on dense hydrous Mg-silicate melts were derived by Monte Carlo simulation method [4] in the systems Mg/Si=1.0, 1.5 and 2.0. In those S(Q)s, first sharp diffraction peaks (FSDP), which may be related to SiO_4 tetrahedral network in silicate melts, shift to higher Q with increasing MgO content and pressure. At ~ 6 GPa, FSDP on Mg/Si=2 melt is 4% higher than that of Mg/Si=1. This indicates that MgO bearing melt is more depolymerized. Pressure dependence on the shifts of FSDP was quite large on the Mg/Si=1.0 system, and the value at ~6 GPa is 18% larger than that of ambient condition[5]. This large contraction can be explained by the shrink of -Si-O-Si- chain-network structure on compression. The results of Fourier transformation of S(Q) show that Si-O inter-atomic distance (nearest neighbor distance in silicate melts) is slightly longer than the dry melt in the Mg/Si=1.0 system (1.64 Å at 3 GPa and 1.65 Å at 6 GPa). This extension may attribute to the bonding balance of Si-O-H (repulsion between Si^{4+} and H^+ and/ or strong O-H bonding). In addition, clear increase of the bonding distance with pressure may show the gradual coordination change of 4-fold to 6-fold Si.

References

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