

Change of water dissolution into magnesium silicate melts at high pressure: Evidence from high-pressure X-ray diffraction study

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Introduction

Structure of silicate melt under high pressure gives us knowledge about magma activity in the Earth's interior, because the physicochemical properties are dominated by the microscopic structures. Since water can be present at mantle edge where magmas are produced, the presence of water may affect magma generation and its physicochemical properties. It is well known experimentally that the composition of magma generated in the shallower parts of mantle is enriched in SiO₂ under hydrous condition. From the microscopic point of view, this phenomenon can be explained from the segmentation of network structure of SiO₄ tetrahedron by H₂O. In hydrous silicate glasses whose structures are considered to be similar to those of hydrous melts, the presence of SiOH group has been reported by some previous studies. Recent studies, however, have revealed that the composition of hydrous magma starts to become enriched in basic composition with pressure [1]. In order to interpret this phenomenon from the microscopic point of view, in situ high pressure and high temperature work is required to clarify the structure of hydrous silicate melt.

Experimental method

Mg(OH)₂ and SiO₂ were mixed in order to make the systems Mg₂SiO₄-H₂O, Mg₂SiO₄-MgSiO₃-H₂O, and MgSiO₃-H₂O, whose water contents were 20.4, 18.3 and 15.2 wt%, respectively. High-pressure X-ray diffraction measurement was carried out at AR-NE5C beamline using a white X-ray beam by energy-dispersive method and a MAX80 cubic-type press. The diffraction profiles were collected by a Ge SSD for 8-10 2θ angles between 3° and 25° in order to obtain the data in the wide *Q* range. The anvils made from tungsten carbide alloy with truncated edge lengths of 10, 6, 4 mm were used. The hydrous silicate melts were encapsulated successfully in a single-crystal diamond sleeve of which the both ends were sealed by platinum lids. The detailed illustration of the high-pressure cell assemblage has been introduced in our previous report [2] and [3].

Results and discussion

The high-pressure and temperature X-ray diffractometry of hydrous Mg-silicate melts was carried out up to 6.9 GPa and 1600 °C. We focused on the shifts of the 1st peaks in the interference functions, *Qi(Q)*, which was obtained from the diffraction data using the analytical

program [4]. The peak is so-called First Sharp Diffraction Peak (*FSDP*). The *FSDP* is often related to the intermediate range structure of the melt, which consists of the silicate network (i.e., the position of the peak indicates the size of the intermediate range ordering in silicate melt). In all the compositions, the *FSDPs* moved to higher *Q* drastically up to 2-3 GPa (note that the *Q* has a reciprocal dimension of Å⁻¹). In contrast, at higher pressure region of 3-5 GPa, the *FSDPs* shift to lower-*Q* side despite of the compression. Those behaviors of *FSDPs* were not recognized in anhydrous Mg-silicate melt reported by [5].

As we stated above, the intermediate range structure in silicate melt means the silicate network which consists of the linkage of SiO₄ tetrahedra (e.g., ...-Si-O-Si-...) in the melt. As an interpretation of the behaviours of *FSDPs* in the hydrous Mg-silicate melts, significant decrease of the intermediate structure should be caused up to 2-3 GPa. Whereas, increase in the intermediate range should be also occurred between 3-5 GPa in all compositions of hydrous melts. The present phenomena imply that the hydrous melt is more depolymerized at low-pressure region up to ~3 GPa and polymerized at higher-pressure region at least to ~5 GPa. This mechanism of the polymerization of hydrous silicate melt at high pressure can be expressed as "SiOMg + SiOH ⇒ MgOH + SiOSi". The left-hand side of that relation denotes the mechanism on the depolymerization caused by dissolution of H₂O into silicate melts to form SiOH group. On the other hands, the right-side indicates the re-polymerization mechanism [6] which enhances the construction of Si-O-Si linkage by the change of the preference of OH from Si⁴⁺ to Mg²⁺ at 3-5 GPa. The relation can be also applied to the interpretation of the magma genesis of MgO-rich liquid at higher pressure.

References

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