A study on structure of hydrous silicate melt by X-ray diffraction experiments using synchrotron radiation

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Structure of silicate melt under high pressure gives us knowledges about magma activity in the Earth's interior, because the physicochemical properties are dominated by the microscopic structures. Since water can be present in mantle wedge 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 SiO_2 component under hydrous condition. From the microscopic point of view, this phenomenon can be explained from the segmentation of network structure of SiO_4 tetrahedron by H_2O. Actually, in hydrous silicate glasses whose structures are considered to be similar to those of hydrous melts, the presence of SiOH unit has been reported by some previous studies. Recent studies, however, have revealed that, above 5 GPa, the composition of hydrous magma starts to become enriched in basic composition with pressure. In order to interpret this phenomenon from the microscopic point of view, in situ high pressure and high temperature work should be needed to clarify the structure of hydrous silicate melt above 5 GPa.

Since the structure of glass can be an analogue of that of melt, several studies have been performed to determine structures of hydrous silicate glasses using spectroscopic techniques such as nuclear magnetic resonance (NMR) and Raman spectroscopy. Those methods have been providing much important knowledge on the structure of hydrous magma. The effect of pressure on the structure, however, can not be provided by such analytical studies on hydrous silicate glasses. Although the direct observation on the structure of hydrous silicate melts is necessary in order to accurately understand the structure with pressures, no experimental investigations on the structure of hydrous silicate melts under high pressure and high temperatures have been reported so far. Many experimental difficulties have prevented us from conducting experiment on the direct observation of hydrous silicate melts. In the present study, in order to solve the technical problems, a new enclosing method of hydrous silicate liquid for X-ray diffraction measurement at high pressure and temperature using single crystalline diamond and platinum was developed. Using the present enclosing method, hydrous silicate melts were enclosed stably at high pressure and high temperature during the data collection with high transmission of X-ray. Thus the first structural data on the dense hydrous silicate melts was able to be derived from in situ high-pressure X-ray diffraction study.

As a result of the structural analyses of diffraction patterns of hydrous Mg-silicate melts, significant decrease of the intermediate structure was observed up to 3 GPa. On the other hands, increase in the intermediate range was also observed between 3-5 GPa in all compositions of hydrous melts. Intermediate range structure in silicate melt means the network structure which consists of the linkage of SiO_4 tetrahedrons (e.g., ...-Si-O-Si-...) in silicate melt. The present phenomena can be interpreted as the evidence for the depolymerization at low-pressure region and polymerization at higher-pressure region of the hydrous silicate melt. This mechanism of the polymerization of hydrous silicate melt at high pressure can be expressed as SiOMg + SiOH = MgOH + SiOSi. The left side of that relation shows the mechanism on the depolymerization caused by dissolution of H_2O into silicate melts to form SiOH unit. On the other hands, the right side indicates the re-polymerization mechanism which enhances the construction of Si-O-Si linkage by the change of the preference of OH from Si to Mg at 3-5 GPa. The relation can be also applied to the interpretation of the magma genesis of MgO-rich liquid at higher pressure.