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課題名　Structural evolution of methanol-water mixture under high pressure

共同研究員氏名　中島　陽一
所属・職名　熊本大学大学院先導機構・助教

受入教員　芳野　極

The stability of H-bonded structures in water-alcohol solutions against thermodynamic variables is a key issue from the view point of physics and life sciences because the solutions are the most common liquids and all living organisms are made of H-bonded constructions. The pressure is one of the thermodynamic variables affecting the H-bonded networks. Several studies have been perfumed to investigate the effect of pressure on individual hydrogen bonded liquids such as water, alcohols and their mixtures [e.g. Yamaguchi1998, Soper2000, Katayama2010]. X-ray diffraction measurements on the structure in water were performed up to 17 GPa, which showing that the local structure of water towards simple one with pressure and approaches to a densely packed one around 4 GPa [Katayama2010]. On the other hands, alcohols and their water solutions were studied at a limited pressure range below a half of GPa because of experimental difficulties especially in trapping highly-volatile and –mobile alcohol liquids. In this study, we have developed the experimental technique on X-ray diffraction measurements of the liquids under high pressure and then succeeded to collect X-ray diffraction (XRD) data of water-methanol mixtures up to 1.6 GPa.

We performed high pressure experiments using a multi-anvil apparatus at the beamline BL04B1 of the SPring-8 synchrotron facility. We examined a couple of configurations of the sample container and cell assembly to confirm the ability to keep water-methanol mixture samples under high pressure, which were machined at the IPM, Okayama University. In the earlier stage of this project, we had issues that a liquid sample amount was highly reduced due to the leaking of liquid samples from sample container upon compression. Finally, however, we found a good configuration with a diamond cap sealed by a piston-shape gold lid, which enabled to safely keep liquid samples even after compressed more than 1 GP. Based on the sample state at the present maximum pressure of 1.6 GPa, a further compression to 10 GPa can be achieved with the same cell-assembly configuration.

We obtained XRD data of three different compositions of water-methanol mixtures containing 20, 30 and 40 mol% methanol, up to 1.6 GPa at 300 K. The energy dispersive scattering from the samples was collected, by using a SSD detector on a two theta goniometer, in the range up to 12 degrees of the scattering angle, thus covering a Q-range up to somewhat above 100 nm^{-1}. An empty pressure cell was also measured for a background collection. Now the data analysis including Reverse Monte Carlo (RMC) simulation is under progress. The analysis will give the pressure evolution and compositional effect of the local structure of liquid water-methanol mixtures.