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Intermolecular interactions in deuterated hydrogen hydrate under high pressure

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Hydrogen hydrate is made of hydrogen-bonded water molecules forming cages or frameworks that include hydrogen molecules. A high pressure structure of hydrogen hydrate, a filled ice Ic structure, is maintained to at least 80.3 GPa. The filled ice structures of many other gas-hydrates decompose below 6.5 GPa, thus, the filled ice Ic structure of hydrogen hydrate shows the remarkable stability. It is expected that intermolecular interactions which induce the stability for the filled ice structure might occur. However, the intermolecular interactions in the filled ice structure have not yet been clarified. In this study, high pressure experiments of deuterated hydrogen hydrate were performed, and the phase changes and the vibrational changes of the filled ice structures were examined. And, the results obtained from deuterated hydrogen hydrate were compared with those from H2-H2O hydrogen hydrate. Then, the isotopic effects and intermolecular interactions in the filled ice structure for hydrogen hydrate were examined.
A lever-and-spring type diamond anvil cell was used in the high-pressure experiments. The pressure was measured by ruby fluorescence methods. In the experiments of H2-H2O and H2-D2O systems, the samples were prepared by the reaction between water and supercritical hydrogen fluid. In the case of D2-H2O system, liquid deuterium cooled by liquid helium was loaded. In situ optical microscopy, X-ray diffractometry and Raman spectroscopy were performed to characterize the samples.
Raman spectra of H2-D2O system revealed that D2 and HD molecules existed in the filled ice Ic structure. These results indicated that the replacement between the hydrogen atoms of framework water molecules and those of guest hydrogen molecules might occur. In the XRD measurements, the compressibility of the filled ice Ic structure for H2-D2O system was different with that of H2-H2O system. These isotopic effects observed in the Raman and XRD measurements could be induced by the differences of the intermolecular interactions between H2-H2O system and H2-D2O system.

Keywords: Hydrogen hydrate, High-pressure, DAC, Isotopic effects, Intramolecular interaction