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メタンハイドレートの低温高圧物性
Physical properties of methane hydrate under low temperature and high pressure

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Methane hydrate, called fiery ice, has the potential to become an important energy resource in the future. It is also thought to be an important constituent of icy planets like Neptune and satellites such as Titan, thus making it an important part of planetary science. Methane hydrate is known to exhibit an sI structure at low pressures and room temperature, and transforms to an sH cage structure at approximately 0.8 to 1.0 GPa. A further transformation to a filled-ice Ih structure (FIIhS) occurs at approximately 1.8 to 2.0 GPa, with this structure consisting of an ice framework similar to ice Ih that contains voids filled with methane molecules The guest methane molecules are rotation freely in the ice framework. The FIIhS has been reported to survive up to 86 GPa at room temperature with two phase change. However, phase changes and properties at low temperatures and high pressures have not been studied so far. This study intended to clarify the changes in phase and properties of FIIhS of methane hydrate.

High pressure and low temperature experiments were performed using clamp-type diamond anvil cells and a helium-refrigeration cryostat. The pressure and temperature conditions were 2.0 to 77.0 GPa and 30 to 300 K, respectively. As the initial materials light-water host sample and deuterated-water one were used.

In situ X-ray diffractometry and Raman spectroscopy revealed existence of three phases and the phase boundaries between them. The first phase is guest orientationaly disordered phase, i.e. well-known FIIhS, which exists below 20 GPa, the second one is guest orentaional ordering phase above 20 GPa, and the third one is another guest ordering phase with different ordering manner. The results demonstrate that phase changes of methane hydrate proceed via proceeding of guest orientational ordering. However, it seemed a quite peculiar that the slopes of phase boundary are negative. Another low temperature experiments performed revealed the volume expansion at low temperature below 150- 100 K. The expansion was confirmed not due to non-hydrostatic effect but to be intrinsic by annealing treatment. The peculiar phenomenon were examined considering to host proton ordering.

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