

The 397th Geodynamics Seminar

Exploration of new phases of methane hydrate under high temperature and high pressure

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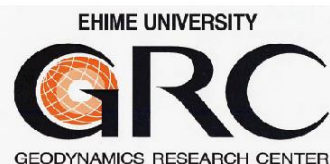
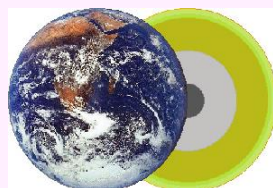
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Abstract

Methane hydrate (MH), called as “fiery ice”, is expected to be a fruitful natural resource. It is also thought to be a major constituent of icy bodies in and outside the solar system. MH is composed of hydrogen-bonded host water molecules forming cages or frameworks that include guest methane molecules. Three phases of MH have been known so far. A low-pressure phase, cubic sI, is stable below 0.8 GPa and it transforms into a hexagonal phase, sH, at 0.8 GPa and further transforms to a orthorhombic filled ice Ih (FIIhS) at 1.8 GPa at room temperature. Recent studies reported that FIIhS transforms to a guest-ordered state phase (GOS phase) at 15- 20 GPa and further to a high-pressure phase (HP1 phase) at 40 GPa. However, the detail structure of HP1 phase has not clarified, moreover, phase changes of MH above 1 Mbar at room temperature and high temperatures have not yet been studied. In the present study, high-pressure experiments were performed to investigate the detail structure of HP1 phase and to explore new phase changes of MH under high temperature and high pressure.

Clamp type and lever-spring type diamond anvil cells were used for high pressure generation. The pressure and temperature conditions in the first stage were from 2.0 to 120 GPa and 300 K, respectively. Pressure measurements were made via a ruby fluorescence method and diamond Raman method. The initial samples of MH were prepared by ice-gas interface method at AIST. The samples were characterized via X-ray diffractometry using synchrotron radiation at BL-10XU, SPring-8, and Raman spectroscopy at GRC, Ehime University.

By annealing the samples via CO₂ laser, in situ X-ray diffractometry identified XRD pattern of single phase of HP1 above 60 GPa, which has not yet been obtained so far. However, the number of the diffraction lines was less only enough to estimate its crystal system, another orthorhombic or tetragonal. Raman spectroscopy showed that a new C-H vibration mode appeared at 31 GPa both for ν_1 and ν_3 , and that the slope of the wavenumber versus pressure clearly changed at around 35 GPa. The changes in C-H modes indicate the change in symmetry of methane molecules, suggesting that the manner of guest orientational ordering in HP1 phase is different from that of GOS phase. Such change in orientational manner of guest methane molecules may result in phase transition to HP1 phase. In the future, high-pressure experiments are going to be performed to investigate new phase changes for MH above 1 Mbar at high temperatures.