Geodynamics Seminar

第365回ジオダイナミクスセミナー

Phase changes of filled ice Ih methane hydrate induced by guest orientational ordering under low temperature and high pressure.

Dr. Hisako Hirai (Distinguished Professor,GRC)

主催:愛媛大学地球深部ダイナミクス研究センター

日時:7/26(金)午後 4時30分~ 場所:総合研究棟 4F 会議室





Abstract

Methane hydrate, also known as "burning ice," is expected to be an environmentally clean energy resource. It is also thought to be a major constituent of icy bodies in and outside the solar system. At relatively low pressures, gas hydrates exhibit clathrate structures comprising host cages formed by hydrogen-bonded water molecules and guest species accommodated within the cages. At higher pressures, several gas hydrates change to filled ice structures, which comprise a host ice framework with the guest species filling in the voids of the framework. The guest molecules are rotating freely in the voids. Comprehensive studies of gas hydrates at room temperature and high pressure have been performed. Yet, phase changes for methane hydrate at low temperature and high pressure above 2.0 GPa have never been reported.

Low-temperature and high-pressure experiments were performed under 2.0-77.0 GPa and 30-300 K using diamond anvil cells and a helium-refrigeration cryostat. In situ X-ray diffractometry revealed distinct changes in the compressibility of the axial ratios of the host framework with pressure. Raman spectroscopy showed a split in the C-H vibration modes of the guest methane molecules. The pressure and temperature conditions at the split of the vibration modes agreed well with those of the compressibility change. The results indicate the following: (i) the orientational ordering of the guest methane molecules from an orientationally disordered state occurred at high pressures and low temperatures; and (ii) this guest ordering led to anisotropic contraction in the host framework. Existing regions of the guest disordered-phase and the guest ordered-phase were roughly estimated by the X-ray study. In addition, above the pressure of the guest-ordered phase, another high-pressure phase developed in the low-temperature region. The deuterated-water host samples were also examined, and the influence of isotopic effects on guest ordering and phase transformation was observed.