

Axis-ratio change induced by guest ordering of filled ice Ih methane hydrate under high pressure and low temperature

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In our previous Raman study, We reported that the orientational ordering of guest methane molecules in a filled ice Ih structure of methane hydrate was observed above 15 to 20GPa. And, by Raman spectroscopy, Sasaki's group reported clear changes in lattice vibration mode of the structure at around 15GPa. That means a certain change in state of the structure. However, by X ray diffractometry, the change in a fundamental structure has not been observed at that pressure range. In this study, the lattice parameters of the filled ice Ih structure were carefully measured at room to low temperature. The results showed that the axis-ratio changed at around 15 GPa, while the fundamental structure was maintained. The similar changes in the axis-ratio were observed for denudated-water methane hydrate. Furthermore, by Raman spectroscopy at low temperature, it is suggested that these changes of the axis-ratio will be induced by the orientational ordering of guest methane molecules in a filled ice Ih structure of methane hydrate.

Keywords: Methane Hydrate, X ray diffraction, high pressure, Raman spectroscopy