

Changes in lattice parameters of filled ice Ih structure of methane hydrate under high pressure

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In our previous Raman study, orientational ordering of guest methane molecules in a filled ice Ih structure of methane hydrate was observed above 15 to 20 GPa. Also, Sasaki's group reported clear change in lattice vibration mode of the structure at around 15 GPa by Raman spectroscopy, showing a certain change in state of the structure. However, change in a fundamental structure has not yet been observed at the pressure range by X ray diffractometry. In this study, the lattice parameters of the filled ice Ih structure were carefully measured at room temperature up to 40 GPa. The results showed that axes ratios changed at around 15 GPa, although the fundamental structure was maintained. The similar changes in the axes ratios were observed at low temperatures and also for denudated-water methane hydrate. The relationship of the axes-ratio change to the orientational ordering of methane molecules in the filled ice Ih structure was discussed.

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