

High-pressure and high-temperature stability of methane hydrate in $\text{H}_2\text{O}-\text{CH}_4-\text{NH}_3$ system

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Methane hydrate is expected to be ubiquitous in space and an important constituent of outer icy planets such as Neptune and Uranus and their moons such as Titan and Europa. The internal structures of these icy bodies have been inferred by spacecraft probes, spectroscopic measurements, and theoretical calculations. The icy mantles of Neptune and Uranus are thought to consist of H_2O , CH_4 , and NH_3 , and methane hydrate is a potential candidate for the major constituent of the mantle. Similarly, recent high-pressure experimental studies (e.g., Bezacier et al., 2014) suggested that the icy crust and mantle of Titan are dominated by methane hydrate, which is likely an essential source of the methane-rich atmosphere at the surface. However, the behavior of methane hydrate in $\text{H}_2\text{O}-\text{CH}_4-\text{NH}_3$ system under high-pressure and high-temperature conditions corresponding to those of icy planetary interiors has not been understood.

Here, we carefully investigated the stability and physical properties of methane hydrate in $\text{H}_2\text{O}-\text{CH}_4-\text{NH}_3$ system under 0.2-6 GPa and 298-413 K using in-situ Raman spectroscopy and X-ray diffraction combined with externally heated diamond anvil cell. Prior to high-pressure and high-temperature experiments, the typical C-H vibration modes of methane hydrate and their pressure dependence were measured at room temperature using Raman spectroscopy to make a clear discrimination between methane hydrate and solid methane which forms through the decomposition of methane hydrate at high temperature. The sequential in-situ Raman spectroscopy revealed that the decomposition temperatures of methane hydrate moderately increased with increasing pressure; e.g., the decomposition occurred at 328 K, 333 K, and 373 K at 0.58 GPa, 1.56 GPa, and 5.18 GPa, respectively. The result obtained by our experiments suggests that methane hydrate can be an important candidate for major constituents of the icy crust of Titan, and has potential to be an essential source of the methane-rich atmosphere at the surface.