

Experimental study on the stability and physicochemical behavior of methane hydrate under high pressure and high temperature

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Methane hydrate are thought to be an important constituent of icy bodies and their satellites, such as Neptune, Uranus and Titan. It is a clathrate compound composed of hydrogen-bonded water cages (host) and molecules or atoms (guests) included in the cages. Methane hydrate has an sI cage structure at low (< 0.8 GPa) pressures and room temperature. It transforms to an sH cage structure at approximately 0.8 GPa, which further transforms to a filled-ice Ih structure at approximately 1.8 GPa. The Ih structure consists of an ice framework similar to ice Ih and voids that are filled with methane molecules (e.g. Loveday et al. 2001; Shimizu et al. 2002). This structure was found to be stable up to at least 86 GPa, supporting that methane hydrate may be stable in the deep interior of icy bodies. Although the sequence of the phase transitions with pressure have been studied well at room temperature, there are only a few studies that addressed the stability of methane hydrate under high pressure and high temperature (Kurnosov et al., 2006; Bezacier et al., 2014). In addition, the pressure range of these previous studies is only limited to < 5 GPa. Therefore, a further investigation is needed to understand the stability and physicochemical behavior of methane hydrate under extreme conditions corresponding to the interior of icy bodies.

In this study, we carefully investigated the stability and decomposition mechanism of methane hydrate in an externally-heated diamond anvil cell in the range of 2-51 GPa and 298-653 K using in-situ Raman spectroscopy and X-ray diffraction. The results show that methane hydrate decomposes to ice VII and solid methane at temperatures considerably lower than the melting curves of solid methane and ice VII in the pressure range of 2-51 GPa. The decomposition conditions of methane hydrate that were obtained at high pressure may help in the modeling of the accretion process and evolutions of icy bodies.

Keywords: methane hydrate, high pressure and high temperature, gas hydrate, diamond anvil cell