

Effect of salinity on self-preservation effect of mixed gas hydrate

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Self-preservation occurs at the dissociation process of methane hydrate and stores the crystal itself even in unstable P-T conditions. We reported in the last JpGU meeting that mixed-gas hydrate composed of methane and ethane showed complicated pattern of dissociation and speed of dissociation depended on both the composition of mixed-gas and the crystallographic structure (Hase *et al.*, 2019). In natural condition, gas hydrates exist mainly in sea-bottom sediments where salinity is abundant. Recently, Prasad and Kiran (2019) reported self-preservation of methane hydrate with NaCl; however, natural gas hydrates comprise not only methane, but also ethane, propane, butane, hydrogen sulfide, etc. In this study, we tried to check preservation effect of mixed-gas hydrate composed of methane and ethane in the presence of salinity.

Fine ice balls were formed from filtered sea water using a spray system and liquid nitrogen, and put 0.7 g into a pressure cell (volume: 30 mL). Then, mixed-gas of methane and ethane was introduced in the cell and their hydrate were formed at 273.2 K. The gas hydrate sample was degassed and recovered at the temperature of liquid nitrogen. We measured compositions of methane and ethane by a gas chromatograph (GC-2014, Shimadzu). Mixed-gas hydrate samples were placed on a temperature-controlled device and kept at 80 K. Raman spectra of hydrate samples were obtained using a Raman spectrometer (RMP-210, JASCO). We observed the Raman spectra of C-H stretching mode at 83 K, and then increased the sample temperature every 10 K until complete dissociation. We calculated the Raman peak area for C-H stretching mode of methane and ethane around 2900 cm⁻¹ by a peak fitting method and obtained the survival rate of gas hydrate samples with temperature.

In the case of pure methane hydrate (composition of ethane: 0%), the survival rate rapidly decreased at around 253 K, agreed with the report by Prasad and Kiran (2019). Although ethane-rich gas hydrate of the structure I showed weak self-preservation at 253 K in the case of no salinity as we reported (Hase *et al.*, 2019), the survival rate also decreased with salinity, indicating that the existence of salt effected the dissociation process of mixed-gas hydrate.

References

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