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Dissociation heat of mixed-gas hydrate composed of methane and propane

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Propane is a former of structure II gas hydrate and found as a major component of guest gas of the natural gas hydrate retrieved at Gulf of Mexico, etc. Enormous amount of latent heat generates/absorbs at the formation/dissociation processes of gas hydrates and controls their thermal condition themselves. Although Handa (1986) obtained the dissociation heat of pure methane/propane hydrate, little attention has been given to their mixed-gas hydrate. We reported dissociation heat of mixed-gas hydrate composed of methane and ethane (Hachikubo et al., 2009). In this study we expanded investigation for the effect of propane on dissociation heat of the mixed-gas hydrate composed of methane and propane.
Samples of mixed-gas hydrate were formed in a pressure cell with an agitation system. 3g of pure water was put into the cell and pressurized by the appropriate composition of the mixed-gas at 27 4K. After well agitation of gas and water, a nucleation occurred by an artificial vibration. The residual gas and the hydrate were sampled and their gas compositions were determined by a gas chromatograph (Shimadzu GC-14B). Propane in the hydrate phase was enriched from the gas phase. The hydrate samples were ground up well in liquid nitrogen and then put into a Tian-Calvet type heat-flow calorimeter (Setaram BT2.15) and their dissociation heat were measured. Inside the calorimeter was pre-cooled at 93K. The internal pressure, temperature and a heat flow to/from the sample were monitored. The sample was heated from 93K to 298K at the rate 0.15 K $min^{1}$to dissociate gas hydrate. Dissociation heat was calculated by an integration of the peak of heat flow and the amount of dissociated gas.
Because propane molecules can be encaged only in the large cages of structure II, it is reasonable to say that methane and propane mixed-gas hydrate primarily forms structure II. Dissociation heat of the mixed-gas hydrate was within the range between those of pure methane and propane hydrates and increased linearly with propane concentration, though a gap between structure I and II was expected in the dissociation heat.

Hachikubo A, Kida M, Okuda M, Sakagami H, Shoji H (2009) Dissociation heat of mixed-gas hydrate composed of methane and ethane. Seppyo 71(5): 341-351

Handa YP (1986) Compositions, enthalpies of dissociation, and heat capacities in the range 85 to 2 70 K for clathrate hydrate of methane, ethane, and propane, and enthalpy of dissociation of isobutane hydrate, as determined by a heat-flow calorimeter. J Chem Thermodyn 18: 915-921

Keywords: gas hydrate, dissociation heat, methane, propane