

Direct measurement of hydration number of gas hydrates

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Hydration number of gas hydrates is an important parameter, and it decides quality of natural gas hydrates. In the case of full occupancy of methane, hydration number of the crystallographic structure I becomes 5.75, however, hydration number of methane hydrate has been reported as 6, indicating that empty cages exist. Hydration number depends on temperature and pressure at the formation process (Cady, 1981; 1983) as van der Waals & Platteeuw model predicts. In this study, we tried to obtain hydration number of gas hydrates directly by a gravimetric determination. We selected argon as a guest gas, forms the crystallographic structure II. Theoretical value of hydration number is 5.67 in the case of full occupancy. We put 5 g of fine ice powder into a pressure chamber (volume: 42 mL), and introduced pure argon (purity: 99.999%, Air Water Inc.). Temperature was controlled at around 273 K to form argon hydrate. After completion of formation, the residual gas was evacuated at 77 K without dissociation of hydrate. Then, we weighed the cell before and after degasification and calculated weights of gas and water. Purity of the argon hydrate sample was checked by PXRD and it is confirmed that contamination of ice was less than 1%. Besides this, argon hydrate was synthesized in another cell (volume: 30 mL) and measured its equilibrium pressure from 270 K to 276 K. Hydration number at the quadruple point was estimated using Clausius-Clapeyron equation. Hydration number of argon hydrate obtained by gravimetric determination decreased with increasing pressure at the formation process; 6.7 at 10.2 MPa and 5.8 at 18.7 MPa. Hydration number at the quadruple point (273.2 K and 8.27 MPa) was 6.9, agrees with the data by gravimetric determination.

References

Cady GH (1981) Composition of clathrate gas hydrates of CHClF_2 , CCl_3F , Cl_2 , ClO_3F , H_2S , and SF_6 . J Phys Chem 85: 3225-3230

Cady GH (1983) Composition of Clathrate Gas Hydrates of H_2S , Xe, SO_2 , Cl_2 , CH_3Cl , CH_3Br , CHClF_2 , CCl_2F_2 , and C_3H_8 . J Phys Chem 87: 4437-4441

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