Phase equilibrium and formation speeds of methane and carbon dioxide hydrate below freezing

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Naturally occurred gas hydrates stored in marine sediments and permafrost have been regarded as a great reserve of carbon. In permafrost area the pressure condition, which affects the stability of hydrate, is complicated since the hydrate crystals are fixed in ice or frozen soil. Methane and carbon dioxide are found in natural gas hydrates. In this study, experiments of gas hydrate formation were conducted below freezing to obtain phase equilibrium data of methane, carbon dioxide and ice system. Besides, formation speeds of mixture gas hydrates were also obtained to understand the kinetic behavior and the controlling factors for the formation of gas hydrates.

Samples of gas hydrate formed from ice powder and guest gases (methane and carbon dioxide) in a pressure cell by monitoring pressure and temperature variations. Temperature was controlled by a liquid bath and kept constant at each experiment in the range of 260-273K. Formation speeds were calculated from the change of the internal pressure. After a gas supply the pressure decreased and became close to the equilibrium value due to a formation of gas hydrate. Phase equilibrium data for methane, carbon dioxide and their mixture gas hydrates were obtained and gas composition was measured by gas chromatograph.

We found that formation speeds of gas hydrates were not controlled only by the temperature and pressure conditions. Formation speeds of carbon dioxide hydrate were much faster than those of methane hydrate in the temperature range of 260-273K. Carbon dioxide gas may affect the surface of both ice and hydrate just below the melting point of ice because carbon dioxide can dissolve more than methane.