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Inclusion of ammonium ion into clathrate hydrate in subsurface ocean of icy moons

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Clathrate hydrates are crystalline inclusion compounds composed of hydrogen-bonded water cages which incorporate hydrophobic gases called guest molecules. The clathrate hydrate could exist not only in the Earth but also in icy moons. One of the most likely candidate is Saturn's icy moon Enceladus, having a global ocean beneath the icy shell. INMS (Ion and Neutral Mass Spectrometer) aboard the Cassini spacecraft has investigated composition of the plumes that includes H2O (< 90 %), CH4, CO2, NH3, and other various organic materials. These results could reflect the composition of the subsurface ocean and imply the presence of clathrate hydrates in the ocean. Bouquet et al. (2015) suggests that the clathrate hydrate should be stable in the subsurface region deeper than 22 km. The clathrate hydrate formation in the ocean should affect concentrations of the ocean components by inclusion of guest and host molecules into clathrate structure. Here we especially focused the effect of inclusion of ammonium ion into clathrate hydrate on ammonium ion concentration in the ocean. Ammonium ion can be incorporated in the clathrate lattice by replacing a part of water cages. To evaluate the ammonium ion inclusion phenomenon in the subsurface ocean, we experimentally investigated the amount of ammonium ions included into clathrate hydrate when hydrate formed in ammonium salts aqueous solution.
As an analogue of CH4 and CO2 hydrate, tetrahydrofuran (THF) hydrate was synthesized in the ammonium chloride solution and the concentration of ammonium ion in the hydrate (Cs mmol/L) was measured. To assess the ammonium ion inclusion, K value which is ratio of Cs to initial concentration (C0 mmol/L) was determined. The K value was about 0.28 when C0 ≤ 10 mmol/L, which was nearly three times higher than that of ice (=0.11). This K value about THF hydrate might be attributed to not only grain boundaries but also replacement with clathrate water cage because the K value was larger than that of ice. This result suggests that excluding ammonium ion from the ocean during clathrate formation is more effective than ice formation.

Significance statement

We experimentally investigated the amount of ammonium ions included into clathrate hydrate when hydrate formed in ammonium salts aqueous solution. The ratio of concentration of ammonium ion in the hydrate to initial concentration was three times higher than that of ice.

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