Discovery of a gaseous sH hydrate former through phase equilibria and structure identification

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One of the way to separate F-gases, which are widely used in semiconductor industries and refrigeration systems while being greenhouse gases, is gas hydrate formation. In this study, c-C_4F_8 (GWP: 8,700, lifetime: 3,200 years) was selected to form gas hydrate in presence of CH_4, since c-C_4F_8 is too large to form gas hydrate by itself (7.66Å). The three-phase (H-L_W-V) equilibrium curve of c-C_4F_8 (5%) + CH_4 (95%) hydrate was moved toward thermodynamically more stable regions compared to that of pure CH4 hydrate, implying the enclathration of c-C_4F_8 and thereby possible structural transformation. Then, PXRD and ^{13}C NNR spectroscopy revealed that both sI (*Pm3n*) hydrate formed by pure CH_4 and sH (P6/mmm) hydrate formed by c-C_4F_8 and CH_4 are coexisting. From these results, it was verified that c-C_4F_8 molecule acted as an sH hydrate former. Also, it was confirmed from the equilibrium curve shift depending on c-C_4F_8 concentrations (c-C_4F_8 2.0% and 5.0%) that c-C_4F_8 is clearly a gaseous guest of sH hydrates. The gaseous sH hydrate former has never been discovered before, and since it is one of F-gases, these results will give a significant impact on gas hydrate application studies.