

## Structural transition induced by CH<sub>4</sub> enclathration occurring in amine hydrate systems

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Two isomers of C<sub>2</sub>H<sub>7</sub>N, dimethylamine (DMA) and ethylamine (EA), are known to be clathrate hydrate formers by themselves. Here, we introduced methane gas as a help gas into both dimethylamine and ethylamine clathrate hydrates and identified their structural transitions using powdered X-ray diffraction (XRD) and solid-state nuclear magnetic resonance (NMR). We observed the structural transitions of amine clathrate hydrates from expanded structure I (cubic Pm3n) to structure II (Cubic Fd3m). In addition, from experimental results obtained through neutron powder diffraction (NPD) and XRD, we found that neither temperature nor pressure affected the hydrate structural transition. Raman spectroscopy was used to identify structural transition occurring in these amine clathrate hydrate systems. In addition, we measured the hydrate equilibrium conditions for amine-water-methane hydrates. The DMA/EA acts as hydrate inhibitors in DMA/EA + H<sub>2</sub>O + CH<sub>4</sub> hydrate systems compared with pure methane hydrate.