

## Structure Transition of Natural Methane Hydrates in Marine Sediments

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In this present work, we offer the clear spectroscopic evidence for the structural transition from sII and sH hydrates to sI hydrate under strong CH<sub>4</sub> atmosphere. The structural transition was confirmed by the analysis of the solid-state NMR spectrum and the Raman spectrum. When replaced by strong external stimulation (CH<sub>4</sub> 110 bar), the mixed CH<sub>4</sub>+C<sub>2</sub>H<sub>6</sub> hydrate of the sII formed by 10 mol% (or 20 mol %) C<sub>2</sub>H<sub>6</sub> was mostly transformed to the sI. Simultaneously, at the same external condition, the formed sH hydrates, methylcyclohexane (or isopentane) + CH<sub>4</sub> + water system, did not show complete structural transition from sH to sI, but were mostly changed to dominant sI hydrate. From these results, the structural transition of initial formed hydrates according to external condition can present the important possibilities to clarify the hydrate structural transition having the dominant sI in deep ocean sediment. Furthermore, the present findings could also have important implications for the understanding of complex hydrate behavior of molecular distribution under strong attacks of guest molecules to the existing cages, and be possibly extended to various hydrate-based fields and inclusion chemistries.