

Effect of Interlayer Ions on Methane Hydrate in Na-montmorillonite

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Natural methane hydrates occurring in marine clay sediments exhibit heterogeneous phase behavior with high complexity, particularly in the negatively charged interlayer region. To date, the real clay interlayer effect on natural methane hydrate formation and stability remains still much unanswered. We first examined the chemical shift difference of ²⁷Al, ²⁹Si, and ²³Na between dry clay and clay containing intercalated methane hydrates (MH) in the interlayer. We also measured the solid-state ¹³C MAS NMR spectra of MH in Na-montmorillonite (MMT) and Camontmorillonite (MMT) to reveal abnormal methane popularity established in the course of intercalation and further performed cryo-TEM and XRD analyses to identify the morphology and layered structure of the intercalated methane hydrate. The present findings strongly suggest that the real methane amount contained in natural MH deposits should be reevaluated under consideration of the compositional, structural, and physical characteristics of clay-rich sediments. Furthermore, the intercalated methane hydrate structure should be seriously considered for developing the in situ production technologies of the deep-ocean methane hydrate.