Structure Transition and Swapping Pattern of Clathrate Hydrates Driven by External Guest Molecules

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We report and discuss the structural transition to sI gas hydrate under strong attacks of external CH_4 guest molecules by spectroscopic analysis. ^{13}C High Power Decoupling NMR spectroscopy was used to identify structure change from strong CH_4 atmosphere of the mixed $CH_4+C_2H_6$ hydrate (sII) and hydrocarbon (methylcyclohexane, isopentane) + CH_4 hydrate (sH). These NMR spectra showed that most of the prepared sII and sH hydrates were transformed to methane hydrate of sI under the methane pressure of 110 bar. In consideration of increase of CH_4 generation by biogenic process in sea water, these structural transitions from dominant CH_4 gas are expected to contribute towards the preponderant occurrence of sI natural hydrates in marine sediments.