Optical Interpretation of CH_4–CO_2 exchange under terahertz light for analysis of CH_4 hydrate replacement process

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Guest exchange method for methane recovery from natural gas hydrates has received recent attention since it can serve double duty of carbon dioxide sequestration (CCS) and methane production. While the methane molecules are exchanged with carbon dioxide, the structure of natural gas hydrates (sI) is maintained. In this study, gas hydrates synthesized from different compositions of a mixtures of methane and carbon dioxide are observed under the terahertz light. Each hydrate sample representing the process of replacement was dissociated to analyze the mole fraction of methane and carbon dioxide by gas chromatography. Obtained refractive indices of samples by terahertz time-domain spectroscopy (THz-TDS) are related to the hydrate composition and a newly discovered methodology that can quantify the extent of replacement via optical constant was introduced. Further, a water framework change from sI hydrate to ice using in situ THz-TDS was investigated during increasing temperature. Overall, this study reveals the process of guest exchange and phase transition from gas hydrate to ice by optical properties in terahertz region, and it can offer a powerful tool in gas hydrate production.