Increasing Hydrogen Storage Capacity using the Metastability of Gas Hydrates

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An intrinsic challenge for gas storage in the hydrates is to balance the paradox between the storage capacity and thermodynamic efficiency. Here, we provide the first experimental evidence for increasing gas (H_2 or N_2) storage capacity at relatively mild condition (at 1 MPa and 263.15 K). With intensification of the sintering of SF6 hydrate particles, we observed an abnormal but unique synchronous behavior in Raman intensities of two guest molecules (SF_6 and N_2/H_2) in hydrates consistently and repeatedly; over time, the scattering intensity for the guests (i) increases, (ii) decreases, and (iii) finally reaches the stable level. Without a concentration change of SF_6, this abnormal behavior must arise from the possible changes in the scattering cross section of the molecules, suggesting that N_2/H_2 strongly interacts with SF_6 in the large cages, resulting in a possible co-occupation during the metastable transition. These observations on the metastability of gas hydrate attest the importance of the sintering effect as a barrier to prevent fast gas diffusion for reaching equilibrium, which could have significant implication in increasing overall gas storage in clathrate hydrates.