

-TIME- AND SPACE-DEPENDENT STOICHIOMETRY EVOLUTION OF METHANE HYDRATE FORMATION IN GLASS BEADS

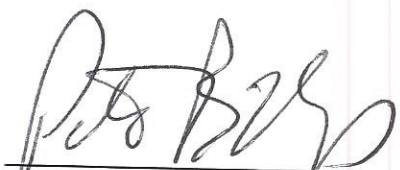
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ABSTRACT

We synthesized methane hydrate in glass beads and used Raman spectroscopy to characterize the pore-scale methane hydrate formation. Analysis of 2D-mapping Raman spectra over 850 hours reveals the dynamic formation process of methane hydrate under natural reservoir conditions (at 17.58 MPa and 280.4 K in 3.5 wt% NaCl aqueous solution).

Contrary to conventional understanding, the thermodynamically stable structure-I methane hydrate does not immediately form. Structure-I methane hydrate has a large- to small-cage molar ratio of 3. In Raman spectra, it is characterized by a large-to-small-cage-peak-area ratio of 3. Raman spectra show that the initially formed metastable methane hydrate was dominated by small cages but poor in large cages, which suggests that small cages form faster than large cages. Over time more large cages gradually formed. After about 500 hours, over 90% of the hydrate in the Raman-mapped area converted to structure-I hydrate.

These results are informative to other laboratory experiments on how to synthesize structure-I methane hydrate to resemble natural methane hydrate reservoirs. These results will also give insight to the molecular-level models of methane hydrate formation and contribute to a better understanding of methane hydrate formation in natural reservoirs.



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