

Crystal-growth studies of natural gas clathrate hydrates using a pressurized optical cell

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ABSTRACT

The crystal-growth behavior of structure I (sI), structure II (sII), and structure H (sH) clathrate hydrates has been studied using a specially designed, pressurized optical cell. Single crystals of each hydrate type, methane sI, methane-propane (95%-5%) sII, and methane-methylcyclopentane sH, were grown in equilibrium with aqueous liquid + vapor ± liquid hydrocarbon. Each structure type exhibits characteristic crystal morphology, which suggests that crystal habit in natural settings, such as sea-floor outcrops, may allow visual identification of hydrate types. In addition, the relative growth rates for different Miller planes for each crystal type were determined. The relative growth-rate schemes and resulting crystal morphology of each structure can be related to the unit-cell density distribution of the small cages in each structure. Four-phase *P-T* equilibrium data for methane-methylcyclopentane sH data were also measured using optical methods. Evaluation of these and previously published phase-equilibrium data for all three known hydrate structures strongly suggests that hydrate assemblages of coexisting sII and sH should be common in natural settings.