

Determination of Local Crystal Orientation of Methane Hydrates in Tailored Confined Media Using Polarized Raman Spectroscopy

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Naturally occurring gas hydrate molecules are energy-rich entities where guest molecules are enclosed in polyhedral host cages of hydrogen-bonded water molecules. Methane hydrates form at high pressure and low temperature. The formation of hydrates is a nucleation and growth phenomenon where molecular-level understanding of chemical physics is of principle importance. Though, in nature, gas hydrates crystallize in the confined media, most reports are available only for the bulk system. Also, the influence of confinement on cage occupancy and different hydrate structure is not well understood. Along these lines, a microfluidic reactor was fabricated with a nano-porous structure placed in the device's channels. The Methane hydrates were crystallized in the nanopores, which act as the confined media, and the orientation of crystals was analyzed using polarized Raman spectroscopy. Using polarized Raman spectroscopy, a detailed methodology is proposed for determining the crystal orientation of the Methane hydrates in the nanopores. Different diameters (20, and 40 nm) of nanopores were used to analyze the confinement effect on the crystal's orientation and cage occupancy. The impact on crystallization in bulk and nanopores was compared based on the stability of crystals.