Contribution ID: **264** Type: **Oral Presentation**

Microfluidic Study of the Pore-Filling and Propagation Behaviors of CH4 and CO2 Hydrates in Porous Media

Monday, 19 May 2025 17:25 (15 minutes)

Gas hydrates are crystalline solids in which guest molecules are trapped within cages formed by water molecules under high-pressure and low-temperature conditions. The phase transition of hydrates, as well as the transport behaviors of the associated gas and liquid phases in porous media, is crucial for CH_4 production and CO_2 storage using hydrates. In this work, we investigate the phase transition behaviors of CH_4 and CO_2 hydrates within porous media under realistic oceanic conditions. To study pore-filling behaviors and spatial stochasticity during hydrate formation in porous media, we developed a low-temperature, high-pressure (LTHP) microfluidic system. This system enables simultaneous visualization of hydrate phase transitions at both the pore and chip scales. It allows for detailed pore-scale morphological studies and observation of collective hydrate formation and propagation behaviors across an area of 45 × 30 mm, etched with over 18,000 pores. To the best of our knowledge, this is the first study of its kind to offer dual-scale imaging over such a large field of view in porous media. Our study reveals density-dependent pore-filling behaviors during gas hydrate formation, addressing knowledge gaps left by earlier microfluidic studies. Hydrates formed from lighter phases, such as gaseous CH4 and CO2, partially fill pores by coating pore walls upon reaching equilibrium. In contrast, hydrates formed from denser phases, like liquid CO2, rapidly cement the pores due to volume expansion, significantly reducing the permeability of the host material. This pore-filling phenomenon was visualized using fluorescence imaging and analyzed through the volume variation index, with kinetics examined under various conditions. We uncovered intrinsic spatial stochasticity in hydrate formation within porous media, demonstrating a random distribution of newly formed hydrates. This randomness can be mitigated through an injection process that simulates CO2 storage, promoting directional hydrate propagation along pressure gradients. Lastly, we propose an alternating CO2-water injection method to enhance CO2 storage capacity and injectivity in shallow seabed environments.

Country

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References

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Session Classification: MS01

Track Classification: (MS01) Porous Media for a Green World: Energy & Climate