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Microfluidic Study of Formation, Dissociation, and Dissolution Dynamics of Gas Hydrates in Porous Media

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Gas hydrates are crystalline solids in which guest molecules are trapped within cages formed by water molecules at high pressure and low temperature. These solids have important applications in natural gas hydrate exploration, CO2 or H2 storage, water desalination, gas separation, and gas/oil transportation. Natural methane (CH4) hydrates are abundant in the seabed sediments and are potential sources for future energy harvesting[1]. On the other hand, carbon dioxide (CO2) hydrates are promising forms of CO2 sequestration due to the large storage capacity. The phase transition of hydrates and the transport behaviors of the relevant gas and liquid phases in porous media are crucial to CH4 production and CO2 storage using hydrates. Therefore, many studies have been conducted on investigating the dynamics of formation, and dissociation of CH4 and CO2 hydrates in porous media using sand or glass bead packs with the help of in-situ imaging methods such as X-ray synchrotron tomography and magnetic resonance imaging (MRI) technology. In this work, we developed a low-temperature and high-pressure microfluidic system for gas hydrate study, allowing for in-situ imaging of the phase transition of hydrates under realistic reservoir conditions of deep seabeds. We studied the formation, dissociation, and dissolution mechanisms of CH4 and CO2 hydrates in both pore scale and chip scale. The hydrates were generated in pure water at 10 MPa and 5 oC subcooling temperature. The dissociation of these two hydrates was induced by decreasing pressure and increasing temperature, respectively. During hydrate formation, we observed the nucleation and propagation of hydrates from the gas-liquid interfaces into the bulk gas, showing various morphologies at the pore scale. The growing kinetics were calculated by analyzing the optical images obtained by a high-resolution camera. Further, we successfully captured the crustal fingering of CH4 gas encased by CH4 hydrates due to the local pressure gradient [2]. From the chip scale, the location of hydrate formation and its propagation in the porous media is stochastic. We found that the induction time for hydrate formation is also stochastic, and the nucleation of hydrate should be triggered by external stimuli such as flow and pressure[3]. During hydrate dissociation, the hydrates remained stable until the pressure or temperature exceeded equilibrium. Then, a drastic transition of hydrates into gases occurs, which results in the fast displacement of gas with liquid in the porous media. In addition, the reformation of hydrates was observed during hydrate dissociation. Finally, we studied the dissolution of CH4 hydrate in undersaturated water and revealed the formation, dissolution, exsolution, and reformation mechanisms of gas hydrates in porous media.

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Primary author: YU, Wei (King Fahd University of Petroleum and Minerals)

Presenter: YU, Wei (King Fahd University of Petroleum and Minerals)

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