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Controlled depressurization of a hydrate-crusted gas capsule: insights from microfluidic experiments and phase-field modeling

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Understanding the evolution of a hydrate-crusted gas capsule during its depressurization is critical to elucidating the fate of methane bubbles escaping from seafloor seeps, a process that controls the impact of seafloor methane leakage on ocean biogeochemistry. While the physics of rising bubbles in a water column has been studied extensively, the process is poorly understood when a hydrate "crust"forms around the gas bubbles during their ascent. Understanding bubble rise, expansion and dissolution under these conditions is essential to determine the fate of bubble plumes of hydrate-forming gases such as methane and carbon dioxide from natural and man-made accidental releases.

In this work, we first present a high-pressure microfluidic experiment to study the controlled expansion of a hydrate-crusted bubble of Xenon in a water-filled and pressurized Hele-Shaw cell of 1 mm thickness. The system is initially pressurized from the Xenon gas inlet (i.e., from the Xe gas bubble) and maintained at a constant pressure of 7.5 MPa and constant temperature of 25 C for 18 hours to ensure equilibrium between Xenon-gas and liquid water. A layer of hydrate shell forms at the gas-liquid interface during this period. At the end of the 18 hours, the gas inlet is disconnected from the system, and the entire cell is depressurized from the surrounding liquid phase at a constant rate of 0.5 MPa/min. During depressurization, the expansion of the gas bubble is controlled by three processes: (1) the volumetric expansion of gas due to changes in pressure; (2) the rupture of the existing hydrate shell that encapsulates the expanding gas phase; and (3) the spontaneous formation of hydrate along the evolving gas-liquid interface. The interplay among these processes results in gas fingering leading to a complex labyrinth pattern, in contrast with the circular gas expansion that would occur in the absence of hydrate formation.

To reproduce the experimental observations, we propose a phase-field model that describes the formation and growth of a hydrate shell on an expanding gas-liquid interface. We design a Gibbs free energy functional for methane–water mixtures that recovers the isobaric temperature–composition phase diagram under thermodynamic equilibrium conditions. The proposed Gibbs free energy is then incorporated into the free energy of the three-phase system (gas-liquid-hydrate) to rigorously account for interfacial effects, phase separation and transformations dynamics: hydrate formation and disappearance, gas dissolution and exsolution. We introduce gas compressibility through a pressure-dependent density formulation. We model the hydrate phase as a highly viscous fluid phase with stress-yielding rheology to reproduce the rupturing behavior of the hydrate layer. We present high-resolution numerical simulations of the model, which illustrate the emergence of complex crustal fingering patterns as a result of gas expansion dynamics modulated by hydrate growth at the interface, as observed in the experiments.

References

X. Fu, L. Cueto-Felgueroso, and R. Juanes. Nonequilibrium thermodynamics of hydrate growth on a gas–liquid interface. Submitted for publication.

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