

Cracking reactive transport processes with gas: Implications for Safe Subsurface Energy Exploitation?

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The interplay between mineral dissolution, precipitation, and gas exsolution plays a crucial role in deep geological repositories for radioactive waste and other subsurface applications such as hydrogen storage and CO₂ sequestration. In nuclear waste disposal, hydrogen gas is generated due to the anoxic corrosion of metallic waste canisters, where iron oxidizes over time. While mineral dissolution and precipitation—where primary minerals dissolve, and secondary minerals form—have been extensively studied, gas exsolution remains poorly understood. Key uncertainties include whether exsolved gases become trapped by mineral precipitates, cause pore clogging, or migrate with fluid flow before dissolving downstream. These gaps limit the accuracy of reactive transport models in predicting long-term corrosion and radionuclide migration.

This study integrates experimental and numerical approaches to (i) determine the reaction and transport conditions leading to gas exsolution and (ii) develop an experimental benchmark at the continuum scale to enhance reactive transport models. We use simplified model systems to study these processes under controlled chemical and hydraulic conditions. Microfluidic experiments, combined with in situ Raman spectroscopy and geochemical modeling, revealed that CO₂ bubbles forming during mineral dissolution serve as nucleation sites for barite. When the barite crystallization rate exceeds CO₂ production, bubbles become enclosed in precipitates. This process is influenced by solution acidity and barite saturation.

To assess gas behavior in porous media, we conducted core-scale experiments. Magnetic Resonance Imaging (MRI) tracked heterogeneous gas production, transport, and relative gas content over time. Additional measurements of pH, pCO₂, differential pressure, and effluent ion concentrations provided insight into reaction dynamics. Scanning electron microscopy revealed reduced porosity due to barite precipitation and localized clogging at bubble surfaces.

Our findings highlight key limitations in existing empirical models such as Van Genuchten-Mualem and Brooks-Corey, which do not account for chemical reactions altering pore structure and wettability. While reactive transport modeling incorporates multiphase flow and mineral reactions, current codes lack full coupling between precipitation, dissolution, and gas dynamics. Advanced mathematical coupling and refined porosity-permeability models are essential for accurately simulating these processes. This framework enhances reactive transport modeling, improving predictions of subsurface integrity in energy and waste storage applications.