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pH-Driven Mixing-Induced Carbonate Mineralization: Insights from Microfluidic Pore-Scale Experiments

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During in situ carbon mineralization, the mixing of injected carbonated water with ambient groundwater can trigger rapid mineral formation. Notably, pH strongly influences carbonate speciation, which is pivotal in driving carbon mineralization. Previous studies have shown that when two fluids mix and create a supersaturated environment with respect to certain minerals, precipitates form at the mixing boundary, resulting in a thin barrier that impedes further mixing over time. However, mineral precipitation can also occur through pH-driven speciation changes during neutralization reactions, particularly when the speciation of mineral constituents depends on pH (e.g., CO₃²⁻). Despite its importance, pH-driven precipitation in the context of pore-scale carbon mineralization has not been investigated. This study investigates the pH-driven carbon mineralization during the mixing of alkaline groundwater and carbonate-rich water.

The microfluidic experimental setup mimics downstream environments in geological carbon sequestration, where the injected carbonate-rich fluid approaches equilibrium with respect to calcite, and OH^- diffusion occurs from the surrounding matrix. To achieve this, we injected a CaCl₂ solution adjusted to pH 11 and a CaHCO₃ solution, slightly undersaturated with respect to calcite at pH 6.5, into separate inlets of a porous microfluidic device using a syringe pump, while maintaining the same Ca^{2+} concentration on both side. The flow rate was varied to assess its impact on the precipitation, with changes in flow rate altering the Peclet number of OH- from 0.19 to 13 and HCO3- from 0.84 to 58. The results revealed that the precipitation band was markedly shifted toward the carbonate-rich side (Figure). As the Peclet number (Pe) increased, the precipitation band narrowed, although it consistently formed on the carbonate-rich side. A parallel experiment mapping pH using a pH-sensitive fluorescent dye revealed pH changes at the interface where the two fluids mixed, aligning with the location of the precipitation band. This shift in the precipitation band is driven by the faster diffusion of OH⁻, which induces deprotonation from HCO₃⁻ to CO₃²⁻. The Grotthuss mechanism explains this rapid diffusion of OH^- ions, distinguishing them from typical solutes dissolved in water. This accelerated diffusion plays a critical role in facilitating the observed shift in precipitation band by enhancing local pH changes and carbonate availability. The corresponding results were modeled using COMSOL, demonstrating good agreement with the experimental observations. This study reveals the importance of considering mixing-driven carbon mineralization especially in the context of pH sensitive reactions. The work has implications to flow channel clogging during mineralization and offers a potential explanation for the formation of veins in geological formations.

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References

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