Coupling Between Flow and Precipitation In Heterogeneous Subsurface Environments and Effects on Contaminant Fate and Transport (Project no. 99272)

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Abstract

This project is aimed at understanding how contaminant transport in heterogeneous porous media is impacted by precipitation and dissolution events through chemical interactions with precipitates and as a consequence of coupling between precipitation and flow. We hypothesize that precipitation/coprecipitation, encapsulation, isolation from flow and alteration of reactive surfaces will contribute to altering contaminant mobility during precipitation events, and that predicting the release of contaminants during precipitate dissolution requires an understanding of how precipitates are distributed and how contaminants are released from the different compartments over time. Using calcium carbonate as a model system, physical experiments and modeling at the pore-scale and continuum-scale will be used to improve the conceptual approach to predicting the impact of flow-precipitation coupling on solute migration. Column and 2-dimensional intermediate-scale experiments with constructed physical and chemical heterogeneities will be used to investigate the movement of fluids and reactive solutes during different types of mixing events that lead to calcium carbonate supersaturation and precipitation. Smoothed particle hydrodynamic modeling will be used to simulate pore-scale mixing and precipitation in heterogeneous porous media and estimate continuum-scale parameters. Continuum-scale modeling will be used to test conceptual models and associated effective parameters that simulate the macroscopic behavior of the experimental domains.

Acknowledgments

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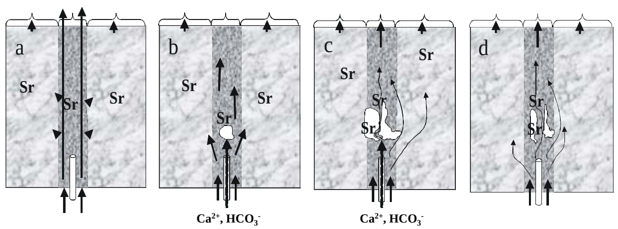
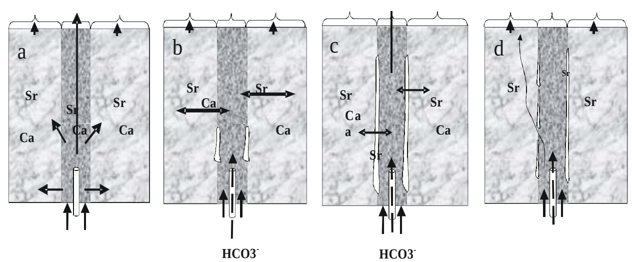
Models for Proposed Experiments

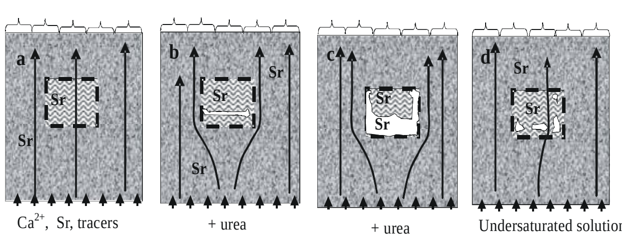
Our goal is to understand how precipitation and dissolution in heterogeneous porous media can impact the short- and long-term transport behavior of reactive metal contaminants. The impact is expected to occur through structured changes in permeability that alter the rate of fluid and solute transport between high and low permeability zones, and through direct interactions between the precipitate and contaminant. Macroscopic, continuum-based modeling of column and intermediate-scale 2-D experiments will be used to characterize, via effective parameters, the averaged transport behavior of fluids and a reactive solute, strontium. Pore-scale modeling and small-scale 2-D experiments will be used to test hypotheses concerning relationships between reaction kinetics, the distribution of precipitates, and solute transport.



Model I: Supersaturated solution will be introduced into a fast flow path bounded by low permeability media. Sr is initially emplaced throughout the system (a). Introduction of supersaturated solution into the fast flow zone is anticipated to cause initiation of precipitation (b). Continued precipitation may cause channeling, and diversion of flow into low permeability zones if constant flux is maintained (c). When undersaturated solutions are subsequently injected, dissolution will occur (d).



Model II: Solutions will be mixed at a high-low permeability boundary. Ca and Sr are initially emplaced throughout the system (a). Following flushing of the fast flow zone with an inert solution, introduction of bicarbonate solution into the fast flow zone is expected to induce precipitation at the high-low permeability boundary (b). As precipitation continues, flux across the boundary is reduced (c). When undersaturated solutions are subsequently injected, dissolution will occur (d).



Model III: Bicarbonate is generated in situ by urea hydrolysis. The central zone contains immobilized urease. Ca and Sr are initially emplaced throughout the cell (a). Following urea introduction, precipitation is expected to start within the urease zone (b). As precipitation continues, flow is diverted around the urease zone (c). When undersaturated solutions are subsequently injected, dissolution is expected to occur (d).

Preliminary Work

- **Precipitation at mixing fronts**
- **Model applications**
- **Analytical methods including x-ray tomography, complex resistivity.**

Figure 1. 2-D cell at INL. Modular inlet and outlet port configurations along all sides allow great flexibility in experimental design.

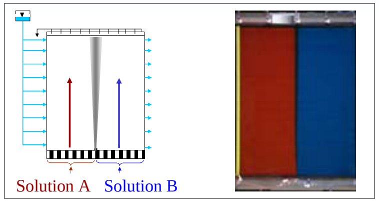


Figure 2. Demonstration of parallel flow, using inert dyes. Mixing occurs at the interface, and the mixing zone widens due to dispersion.

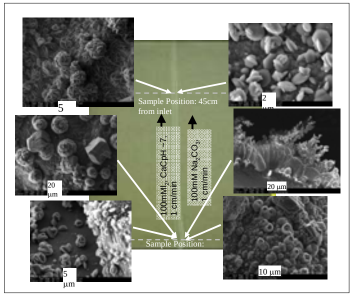


Figure 3. Examples of precipitate morphologies observed at different locations in mixing zone between parallel flows of $CaCl_2$ and Na_2CO_3.

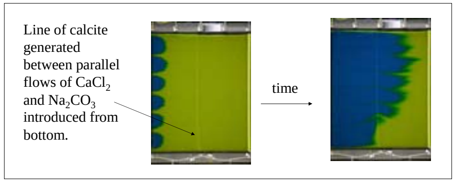


Figure 4. Demonstration of flow impedance due to calcite precipitation. Inert dye injected perpendicular to original parallel flows of reactants (introduced from the bottom). Dye impeded most near start of original mixing zone; farther into the zone, permeability was less affected.

Modeling

Precipitation events will be simulated at the pore scale, but linked to the macroscopic simulations. Figure 5 is an example of a macroscopic simulation of solute mixing under the condition of parallel flows of reactants. It predicts zones of supersaturation wider than the zones where precipitate was actually observed. Figures 6 and 7 are examples of Smoothed Particle Hydrodynamics (SPH) modeling to simulate precipitation associated with the parallel flow case (Figure 6) and also a scenario (akin to Model I) where a supersaturated solution flows through a high permeability channel (Figure 7).

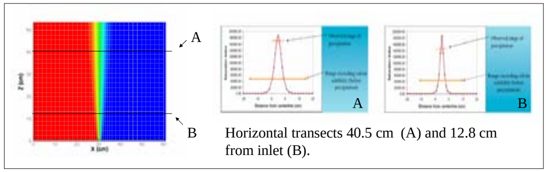


Figure 5. Macroscopic simulation of solute mixing along interface of parallel flows of reactants. Figures on right show calcite saturation conditions for different distances from injection.

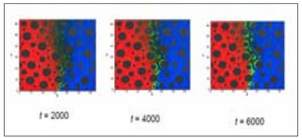


Figure 6. SPH simulation of precipitate formation at interface between parallel flows of reactants, coming from bottom.

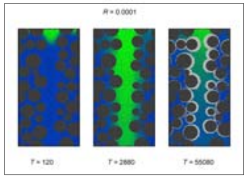


Figure 7. SPH simulation of precipitation due to infiltration (from the top) of a supersaturated solution into a high permeability channel.

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