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Pore Scale Modeling of Calcium Carbonate Reactive Transport

Scientific Achievement

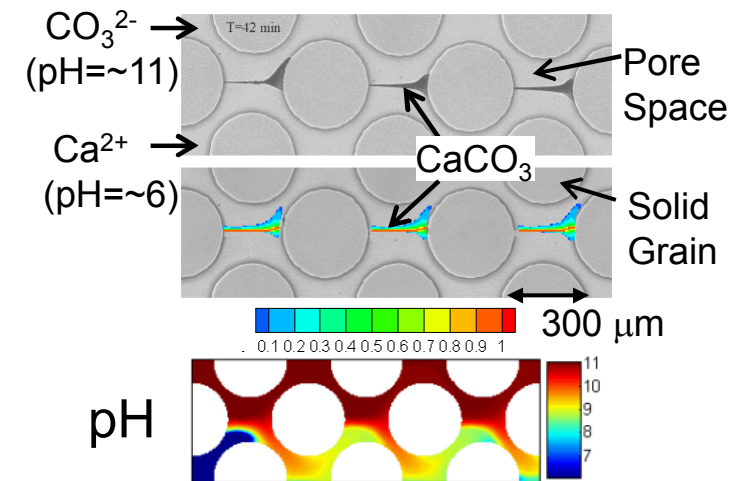
Developed a novel pore scale reactive transport model of coupled fluid flow, reactive transport, and calcium carbonate (CaCO_3) precipitation and dissolution

Significance and Impact

Improved understanding of the fundamental physico-chemical processes of CaCO_3 precipitation and dissolution at micro (pore) scale for coupled reactive transport systems perturbed by geological CO_2 injection

Research Details

- A newly developed pore-scale model is used to simulate transient experimental results of CaCO_3 precipitation and dissolution in a microfluidic pore network (shown in Figure)
- Pore scale model was able to capture the precipitate morphology and precipitation rate
- Proper estimation of the effective diffusion coefficient and the reactive surface area is necessary to adequately simulate precipitation and dissolution rates
- The model developed in this study allows us to simulate and mechanistically evaluate precipitation and dissolution of CaCO_3 observed at pore scale

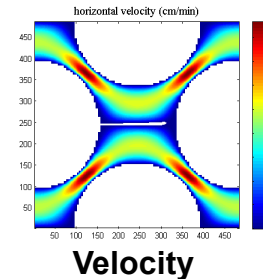


Experimental image of CaCO_3 precipitates (top), simulated CaCO_3 volumetric content (middle), and simulated pH distribution at 42 min. In the microfluidic pore network, precipitation is induced by transverse mixing along the centerline in pore bodies.

H. Yoon, A. J. Valocchi, C. J. Werth, and T. Dewers, Water Resources Research, 48, W02524 (2012)

Pore Scale Model Framework

Lattice Boltzmann Method:
Velocity field (u) at pore scale



Finite Volume Method: Reactive transport at pore scale

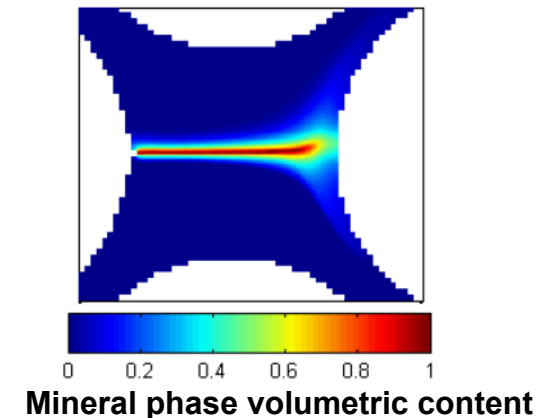
$\Psi_j = C_j + \sum_{i=1}^{N_{eq}} \nu_{ji} C_i$ Chemical equilibrium in bulk fluid (e.g., H^+ , HCO_3^- , ...)
Extended Debye-Hückel Equation for activity coefficients

$$D \frac{\partial \Psi_j}{\partial \mathbf{n}} = -I_m \quad \text{on reactive surface}$$

$$I_m = k_{cc} \left([\Omega]^n - 1 \right)^m \quad \Omega = \frac{Q_{cc}}{K_{sp}} \text{ or } \ln \left(\frac{Q_{cc}}{K_{sp}} \right)$$

Update of $CaCO_3$ volumetric content (V_m)

$$\frac{\partial V_m}{\partial t} = \overline{V}_m s_m k_{cc} \left(\left[\frac{a_{Ca^{2+}} a_{CO_3^{2-}}}{K_{sp}} \right]^n - 1 \right)^m$$



Δt



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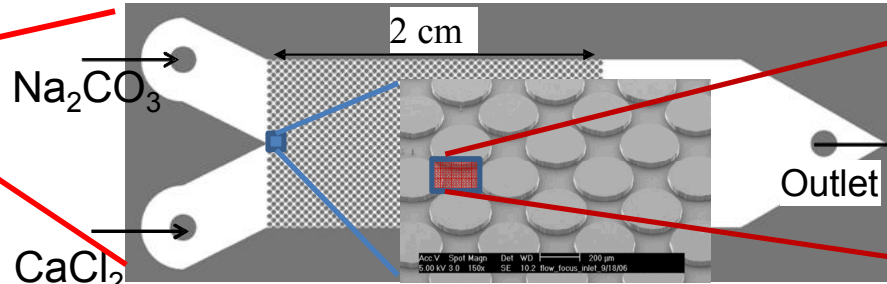


Microfluidic Experiment

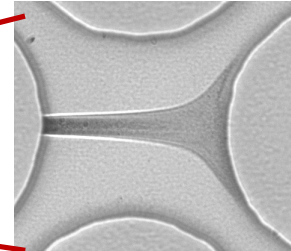
Experiment Setup



3D Laser scanning confocal microscope

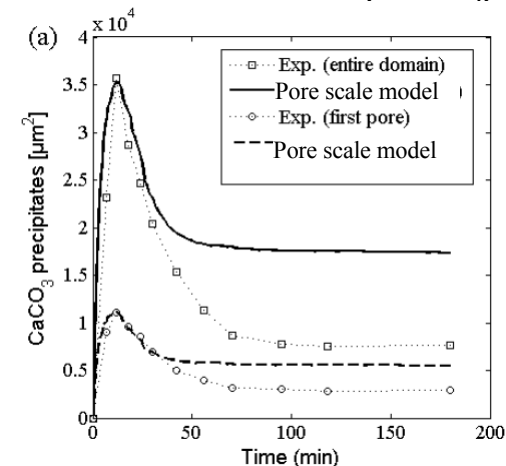


Microfluidic pore network



Microscopic image of calcium carbonate (CaCO_3) precipitates

- Microfluidic pore network (micromodel)
 - 2D representation of porous media etched into Si wafer with any pore geometries at $\sim 0.1 \mu\text{m}$
- Two solutions are mixing along the centerline and reactive product (e.g., CaCO_3) forms
- Flexible with a range of concentrations and solution chemistry
- 2D and 3D in-situ observations with optical or confocal microscope



Comparison of CaCO_3 precipitation area over time. Model captures precipitation and dissolution rates well up to 30 minute, and then under-predicted dissolution rate

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