Effects of physicochemical properties and structural heterogeneity on mineral precipitation and dissolution in saturated porous media

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I. Effects of physicochemical properties on the transport behavior of CaCO$_3$

II. The interplay between structural heterogeneity and reactive transport processes
Challenges

I. How to visualize pore-scale processes during experiments?
II. How to quantify the effects of structural heterogeneity?

Bultreys et al., 2022
Ferreira et al., 2022
Methods: Column experiments

<table>
<thead>
<tr>
<th></th>
<th>S1 mmol</th>
<th>S2 mmol</th>
<th>A1</th>
<th>A2</th>
<th>Q1 ml/min</th>
<th>Q2 ml/min</th>
</tr>
</thead>
<tbody>
<tr>
<td>C1</td>
<td>1.2</td>
<td>1.2</td>
<td>PH=2</td>
<td>PH=11.2</td>
<td>0.4</td>
<td>0.4</td>
</tr>
<tr>
<td></td>
<td>6</td>
<td>6</td>
<td>PH=5</td>
<td></td>
<td>0.6</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>12</td>
<td>12</td>
<td>PH=7</td>
<td></td>
<td>0.8</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td>24</td>
<td>24</td>
<td>PH=9</td>
<td></td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td></td>
<td>60</td>
<td>60</td>
<td>PH=11</td>
<td></td>
<td>1.2</td>
<td>1.2</td>
</tr>
<tr>
<td>C4 - C8</td>
<td>12</td>
<td>12</td>
<td>PH=6.5</td>
<td>PH=11.2</td>
<td>0.6</td>
<td>0.6</td>
</tr>
</tbody>
</table>

Column 1-3: medium particle size quartz sand under different environmental conditions;

Column 4-8: different setups and levels of heterogeneity.
Methods: HYDRUS – PHREEQC simulations

Governing equations

- Richards equation:

\[
C(h) \frac{\partial h}{\partial t} - \nabla \cdot K(h) \nabla h - \frac{\partial K}{\partial z} = 0
\]  \hspace{1cm} (1)

- Convection – diffusion equation:

\[
\frac{\partial C_i}{\partial t} + \vec{u} \cdot \nabla C_i = D \nabla^2 C_i + R
\]  \hspace{1cm} (2)

\[
R = k C_{CaCl_2} C_{Na_2CO_3}
\]  \hspace{1cm} (3)

- Reaction:

\[
Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3
\]  \hspace{1cm} (4)

Simulation processes

- Basic information
- Water flow
- Solute transport
- Boundary condition
- Initial condition
Methods: XCT visualization and post-processing

XCT image processing

Morphological calculation

pore volume

surface area

Euler number

convexity

Blunt et al., 2017; Legland & Carreras, 2018
Results: Transport behavior under different physicochemical conditions

C1: the effects of salinity
Salinity increase → precipitation enrichment → weak Ca\(^{2+}\) breakthroughs

C2: the effects of acidity
Acidity increase → precipitation shrink → strong Ca\(^{2+}\) breakthroughs

C3: the effects of flow rate
Flow rate increase → precipitation shrink → strong Ca\(^{2+}\) breakthroughs
Results: Effects of structural heterogeneity

Breakthrough curves of C4

- Low porosity promotes precipitation; while precipitation decreases pore connectivity.
- The increase of particle size leads to early breakthrough and higher peak concentration.
Results: Effects of structural heterogeneity

Breakthrough curves of C5~8

3D visualization of C5~8

- Vertical layering causes more CaCO₃ precipitation accumulated in the columns.
Results: Downscaling from XCT perspective

Sliced images and histograms

- Significant decrease of pore connectivity and increased level of heterogeneity on grain surface after the PD process.

- CaCO₃ is often precipitated in narrower pores and accumulated first on concave surface.

A: sand, B: pore, and C: carbonate

Surface area | Circularity | Euler number | Convexity
--- | --- | --- | ---
![Graphs for coarse](image)

![Graphs for medium](image)

![Graphs for fine](image)
I. The traditional breakthrough curves were able to demonstrate the macroscopic behavior of reactive transport and reveal the effect of physiochemical properties on precipitation – dissolution process.

II. XCT visualization and morphological calculation provide microscopic information that help explain the macroscopic transport phenomenon in heterogeneous porous media under various conditions.

III. High-quality data from XCT images could serve as an input for further pore-scale modelling and simulations.
Thanks for listening!

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