Pore-Scale Controls of Reactive Transport in Carbonate Porous Media

Wen Song
Hildebrand Department of Petroleum and Geosystems Engineering
University of Texas at Austin

American Geophysical Union Fall Meeting
December 9 - 13, 2019
San Francisco, CA, USA
**Geological Storage**

Atmospheric emission of CO₂ > 40 billion tons/year

**Reactive Transport**

\[ H^+ + CaCO_3 \rightarrow CO_2 + H_2O + Ca^{2+} \]

- CO₂-acidified storage brine
- Modifications to storage reservoir structural integrity

**Wormholing**

- Pathways for leakage
Subsurface Engineering: A Question of Scales

Field Scale
- Top of Structure
- Seal
- Gas
- Water
- Reservoir

Microscopic Scale
- Sandstone Grains
- Oil in Pore Spaces

Reservoir Scale
- Block of Sandstone

Society of Petroleum Engineers
Reactive Transport in Carbonates

Interface  Pore  Pore-Ensemble  Reservoir

300 µm  1 mm  80 m

Graphs showing normalized calcite volume over time with labels for different regimes and conditions.
Interface-Scale Dynamics: Fundamental Mechanism Discovery

**Grain-Engulfment Mechanism**

\[
\text{CaCO}_3(s) + 2\text{H}^+_{(aq)} \rightarrow \text{CO}_2(g) + \text{Ca}^{2+}_{(aq)} + \text{H}_2\text{O}(l)
\]

St $<$ Da  
PeDa $>$ 1  
Two-phase transport-limited reactive transport  
Grain-Engulfment Regime

Acidic fluid  
PDMS channel  
Calcite post  
Produced CO$_2$  
Width $W = 1.5$ mm  
Width of calcite $W_{\text{calcite}} = 500 \mu$m

Injection velocity = 10 m/day  
2% hydrochloric acid  
Outlet pressure = $P_{\text{atm}}$

Normalized Calcite Volume ($\mu$m$^3$ / $\mu$m$^3_{\text{initial}}$)

Time (s)

Steady Dissolution  
Grain Engulfment

[Song et al (2018) Lab on a Chip]
Phase Diagram

\[ St = \frac{1}{Pe} \times (PeDa) \]

Regime I: Single phase
Regime II: Reaction-limited
Regime III: Transport-limited

PeDa = CO₂ reaction / CO₂ diffusion in aqueous phase

[Song et al (2018) Lab on a Chip]
Biogenic Calcite Microfluidics: Geometrically and Mineralogically Representative Model

Sporosarcina pasteurii

[Song et al (2018) Lab on a Chip]
Biogenic Calcite Microfluidics: Geometrically and Mineralogically Representative Model

Cementation Fluid:
- 1M urea
- CaCl₂

[Song et al (2018) Lab on a Chip]
Biogenic Calcite-Functionalization of Micromodel

[Song et al (2018) Lab on a Chip]
Grain Engulfment in Porous Media

- 50 m/day
- 2% HCl injection
- 5 psi, 35°C
- Sped up 60x
- Separate CO2 gas phase evolved
- Protective grain-engulfment effect locally

Sped up 60x

[Song et al (2018) Lab on a Chip]
Grain Engulfment: Pore Scale Dynamics

CO$_2$(g)  Calcite grain
Silicon grain

Sped up 120x

[Song et al (2018) Lab on a Chip]
Grain Engulfment on Local Dissolution

[Image of grains and CO$_2$ with time progression and graph showing normalized calcite volume over time.]

[Song et al (2018) Lab on a Chip]
Reservoir Implications: Far from CO$_2$ Injector

- 50 m/day injection of 2 % HCl
- 1200 psi, 35 °C
- No separate CO$_2$ phase evolution, constant rate of calcite dissolution

Dissolution rate \( \sim 60 \, \mu m^2/s \)

Da \( \sim 7.3e-2 \)

[Song et al (2018) Lab on a Chip]
Reservoir Implications: Close to CO₂ Injector

- 50 m/day injection of 2 % HCl saturated with CO₂
- 1200 psi, 35 °C
- Separate supercritical CO₂ phase evolution, reduced rate of local calcite dissolution

[Song et al (2018) Lab on a Chip]
Reservoir Implications: Close to CO₂ Injector

- 50 m/day injection of 2% HCl saturated with CO₂
- 1200 psi, 35 °C
- Separate supercritical CO₂ phase evolution, reduced rate of local calcite dissolution

[Song et al (2018) Lab on a Chip]
Discovery: Fundamental Mechanism

Engulfed calcite grains

Exposed calcite grains

Acid in preferential flow path

Dissolving calcite grain

Calcite grain

CO₂

[Song et al (2018) Lab on a Chip]
Engineering Science Advances: Micro/Nanofluidics in Energy

INFRASTRUCTURE DEVELOPMENT

• Real-Rock Microfluidics for Fundamental Pore-Scale Understanding

FUNDAMENTAL DISCOVERY

• Grain-Engulfment on Local Reactive Transport

REAL-WORLD IMPACT

• Mechanistic Depiction of Wormholing in Carbonate CO₂ Storage Reservoirs

[Song et al (2018) Lab on a Chip]
Acknowledgement

Anthony R. Kovscek, Martin Ferno, Folake Ogunbanwo