

Pore-Scale Dynamics of Multiphase Reactive Transport in Carbonates under CO₂-Acidified Brine Injection

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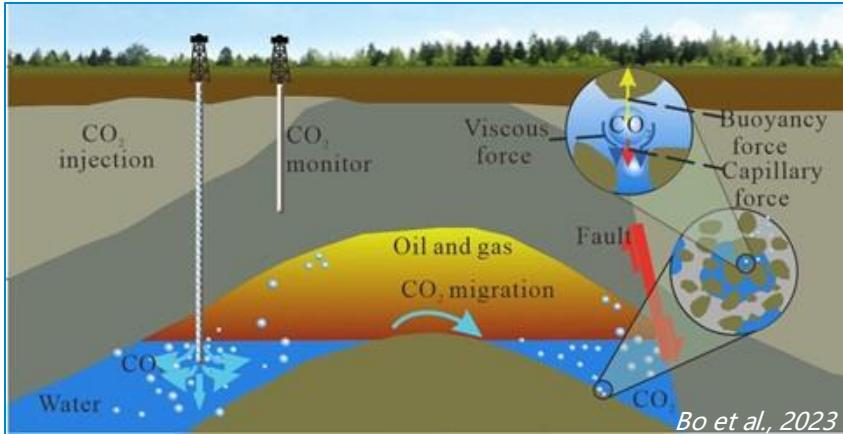
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Outline

1. Overview
2. Objectives: Multiphase Reactive Transport
3. X-ray Tomography Experiments and the Impact of Transport Heterogeneity
4. Observations and Conclusions
5. Future Work

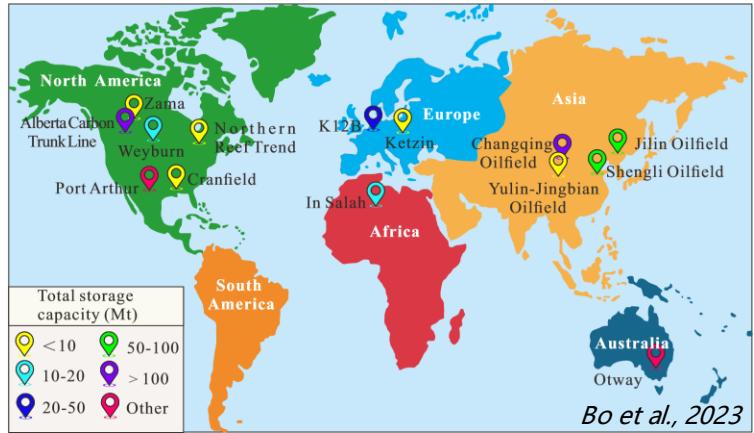
1. Overview

• Research background



Depleted Hydrocarbon Fields

Less additional costs to implement
Mature technical processes
Ease of management

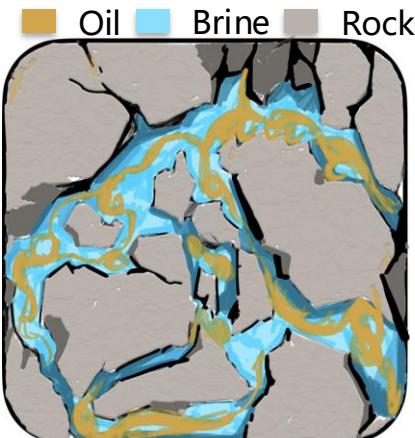


Global CO₂ Storage Projects in Depleted Hydrocarbon Fields

Global CO₂ storage capacity
of 900-1200Gt

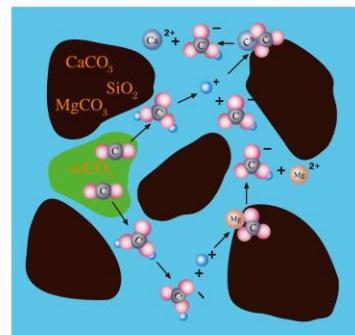
- Challenges

Reactive Transport & Multi-phase flow
 Carbonate rock partially saturated with hydrocarbon phase.

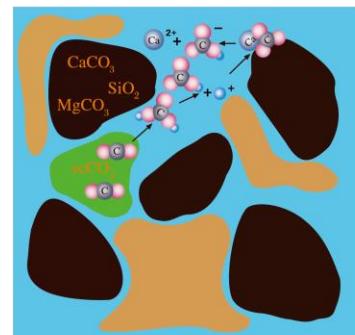


Carbonate rock saturated with oil and brine

Heterogeneity
 Hydrocarbon phase will change the transport heterogeneity and may block the CO₂ equilibrated brine from reacting.

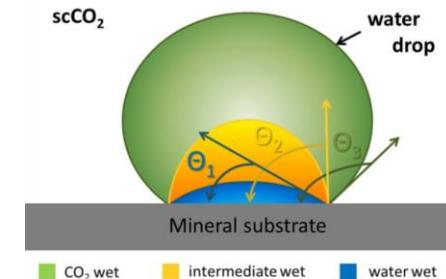


Reactive Transport Phenomena



Reactive Transport in Presence of Oil

■ scCO₂/CO₂(l) ■ brine ■ rock ■ oil



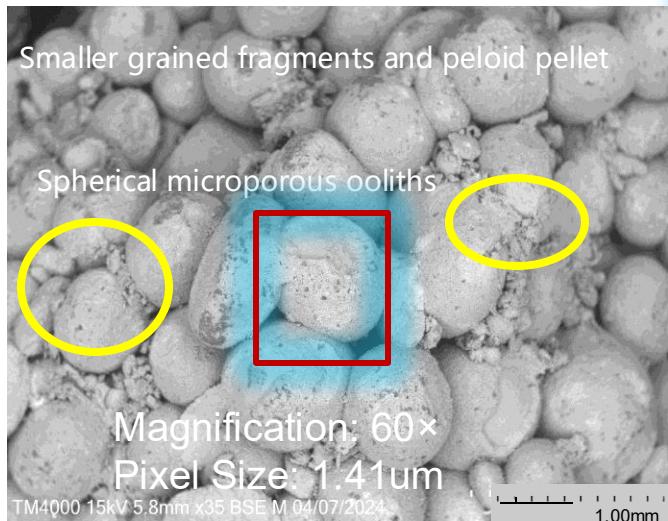
$\Theta_1 \approx 30^\circ \Rightarrow$ strongly water wet system
 $\Theta_1 \approx 90^\circ \Rightarrow$ intermediate wet system
 $\Theta_1 \approx 135^\circ \Rightarrow$ strongly CO₂ wet system

Wang et al., 2024
 Wettability States

2. Objective: Reactive Transport in Multiphase Flow

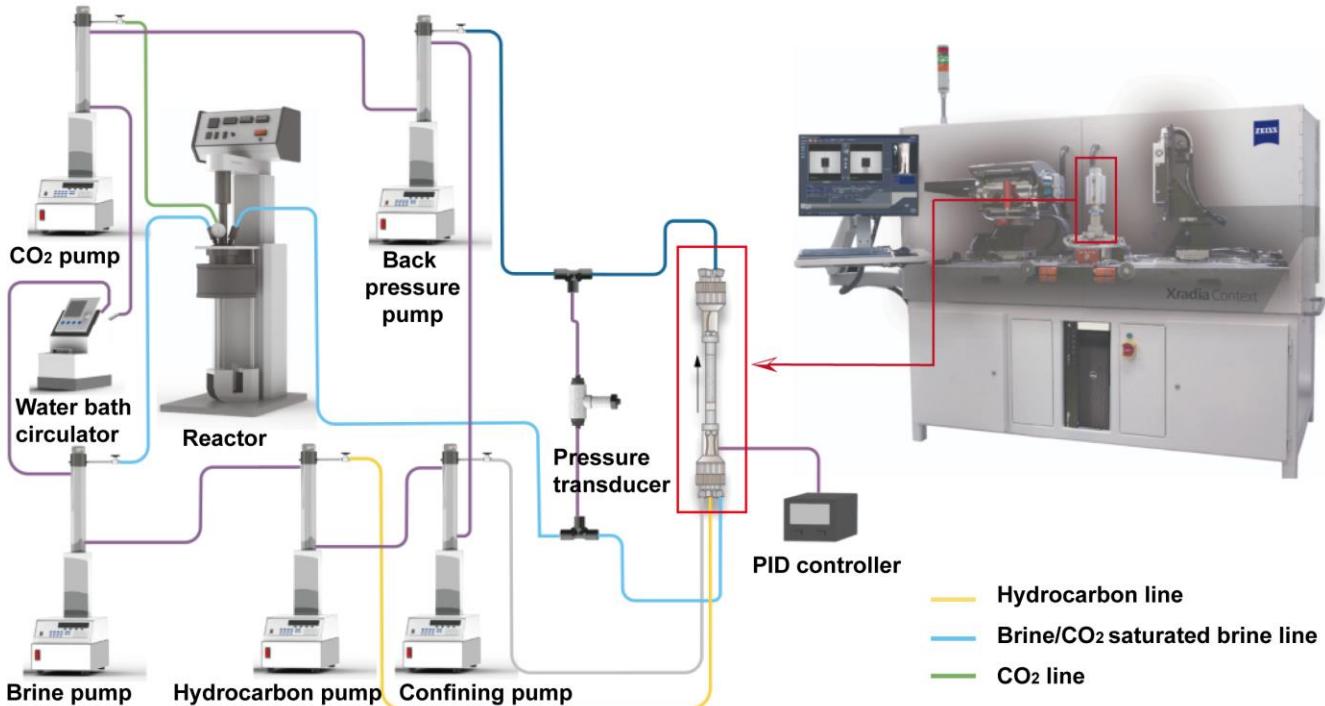
● Objective

- The impact of changes in **transport heterogeneity** caused by **the introduction of hydrocarbon phase** on flow mechanisms and dissolution patterns.
- A permeable Ketton oolitic limestone (99% calcite).



3. X-ray Tomography Experiments and the Impact of Transport Heterogeneity

• Experimental apparatus



Measurement System

1. Fluid Flow Unit

ISCO Pumps
High T&P Reactor
High T&P Core Holder
Back Pressure Regulator

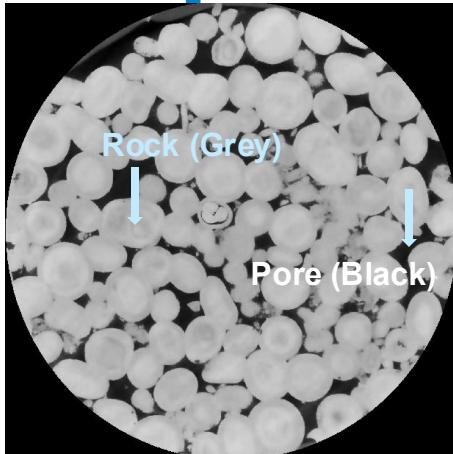
2. Monitoring Unit

X-ray CT
Pressure Transducer

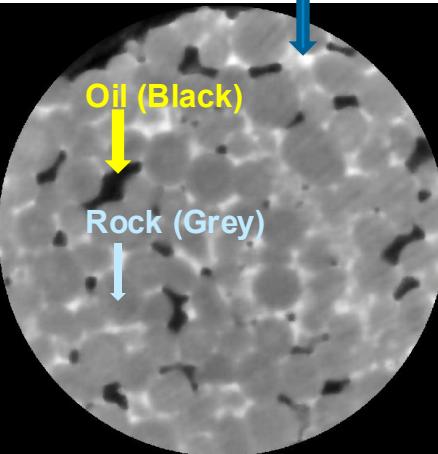
3. Stability Unit

Water Bath Circulator
PID Controller
Insulation Cover for Tubings

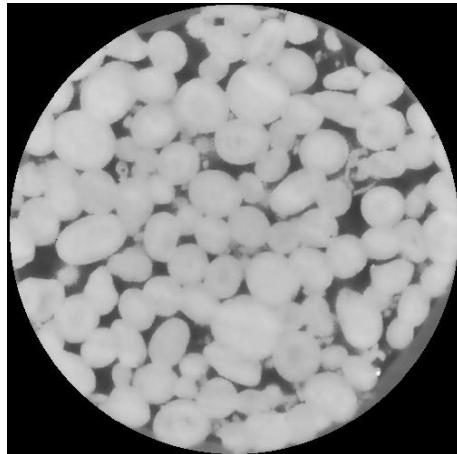
● Samples



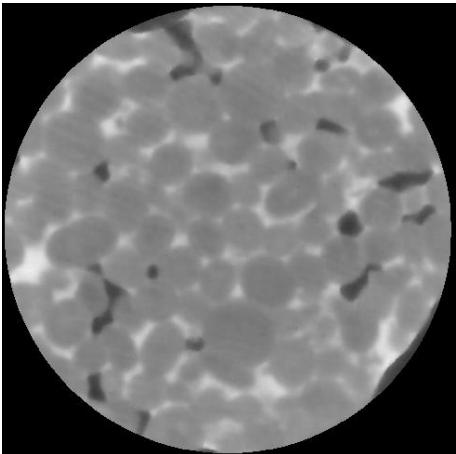
Sample1



Sor= 39.8%



Sample2

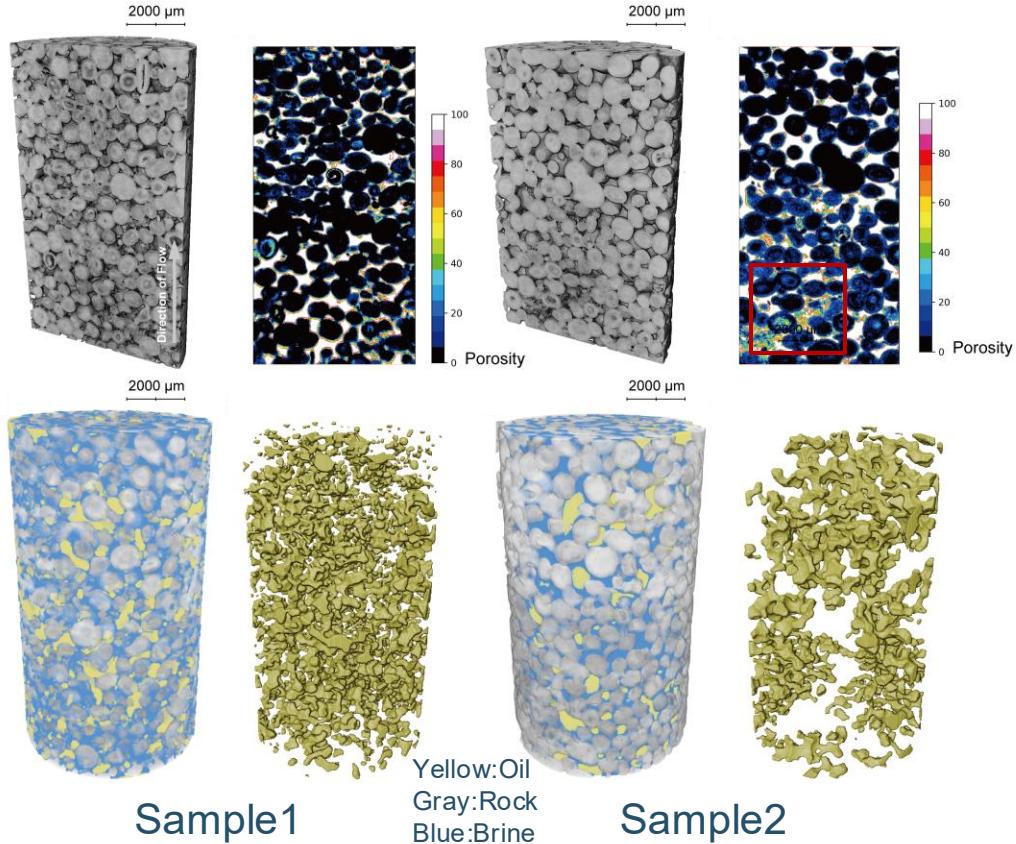


Sor= 31.1%

Rock Sample	S1	S2
Length, mm	120	120
Diameter, mm	60	60
Permeability(Darcy)	2.46 ± 0.10	2.74 ± 0.18
Micro-CT porosity (Marco-Porosity)	0.147	0.163
Oil Saturation	39.8%	31.1%

- Water-wet Ketton limestone saturated with oil and brine in different saturations.
- Quantitatively characterize transport heterogeneity based on their velocity distribution.

● Pore-scale heterogeneity



Dual heterogeneity:

- Pore-scale structural heterogeneity.
- Residual oil distribution.

Sample 1:

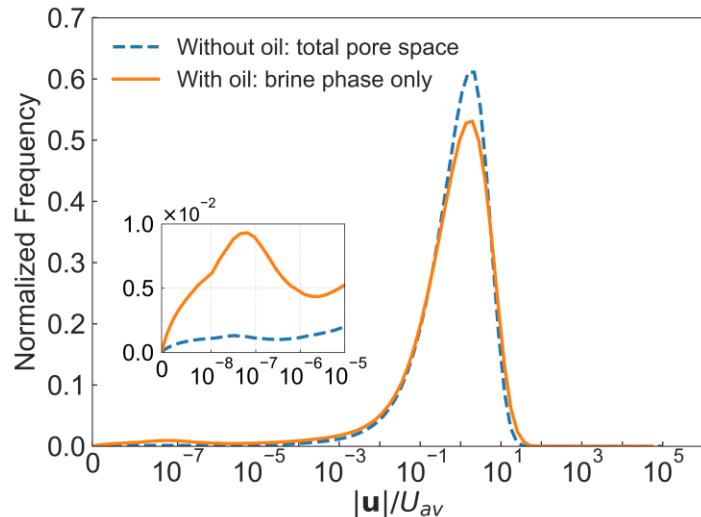
- A well-connected network of intergranular macropores leads to a dispersed distribution of residual oil.

Sample 2

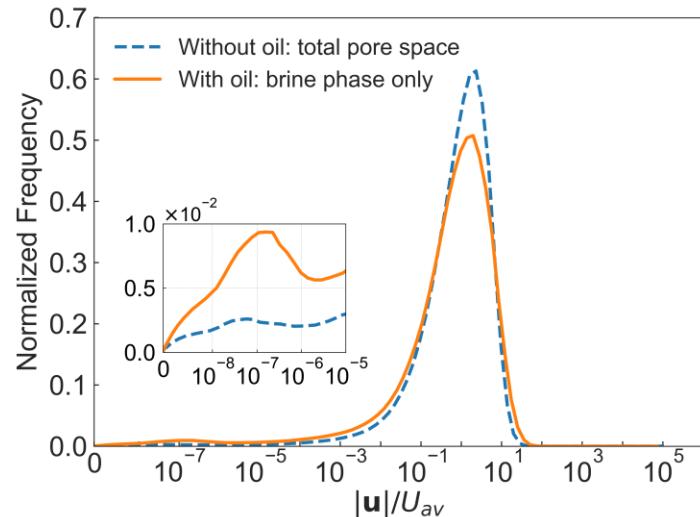
- A dual-porosity system with intergranular macropores and poorly connected intermediate-sized pores leads to localized residual oil clusters.

● Pore-scale heterogeneity

- A finite volume solver implemented in OpenFOAM solves the Navier-Stokes and volume conservation equations simultaneously.
- Obtained voxel velocities U and then calculated probability density functions (PDFs).



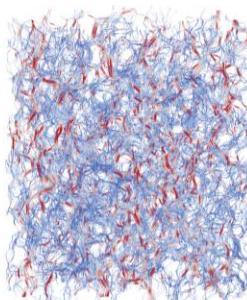
Sample1



Sample2

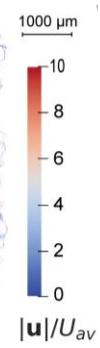
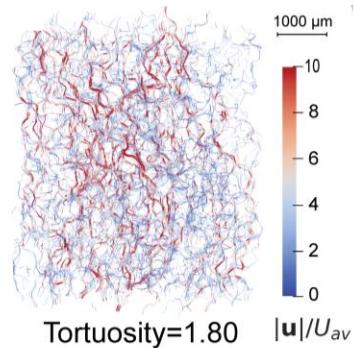
● Pore-scale heterogeneity

Without Oil: total pore space
(Initial velocity distribution)



Tortuosity=1.63

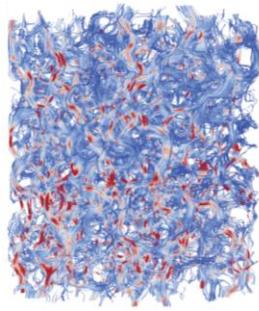
With Oil:
Brine phase only



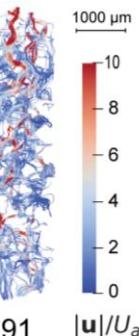
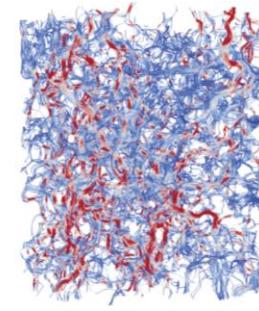
Tortuosity=1.80

Sample1

Sample2



Tortuosity=1.71



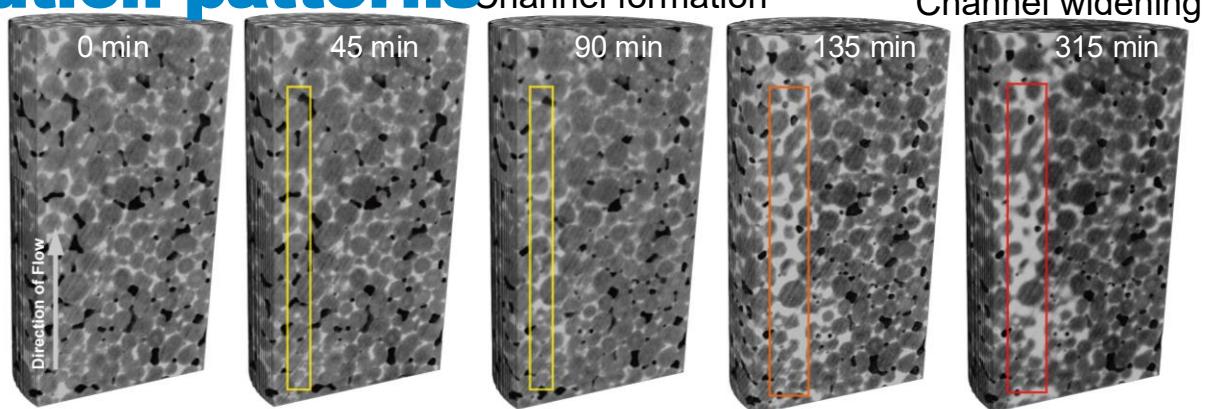
Tortuosity=1.91

Using the tortuosity of streamlines to characterize transport heterogeneity.

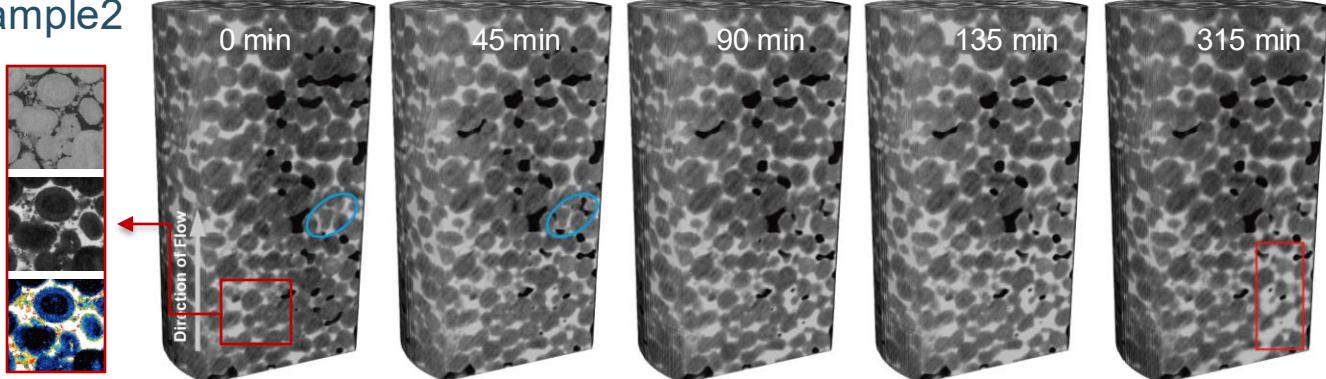
- Lower tortuosity: Flow pathways are simpler and less obstructed.
- Higher tortuosity: Oil restricts brine flow, making pathways more complex.
- Sample 1: Tortuosity increased by 10.4%; Sample 2: Tortuosity increased by 11.6%.

• Dissolution patterns

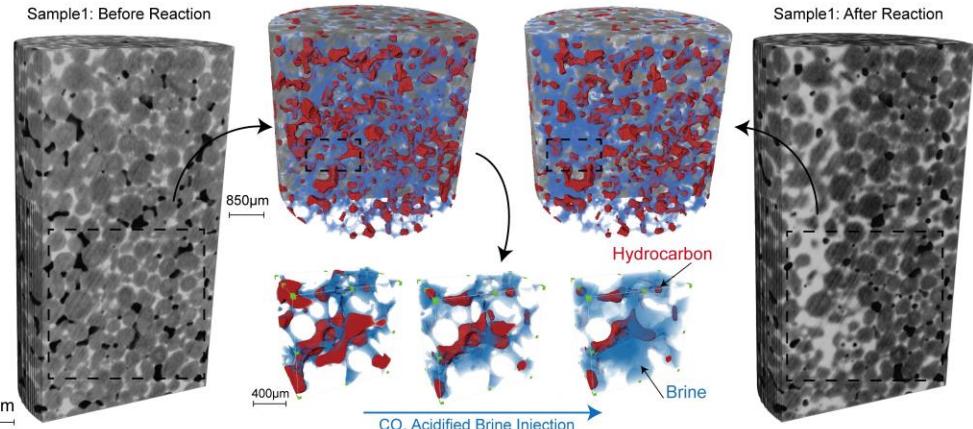
Sample1



Sample2

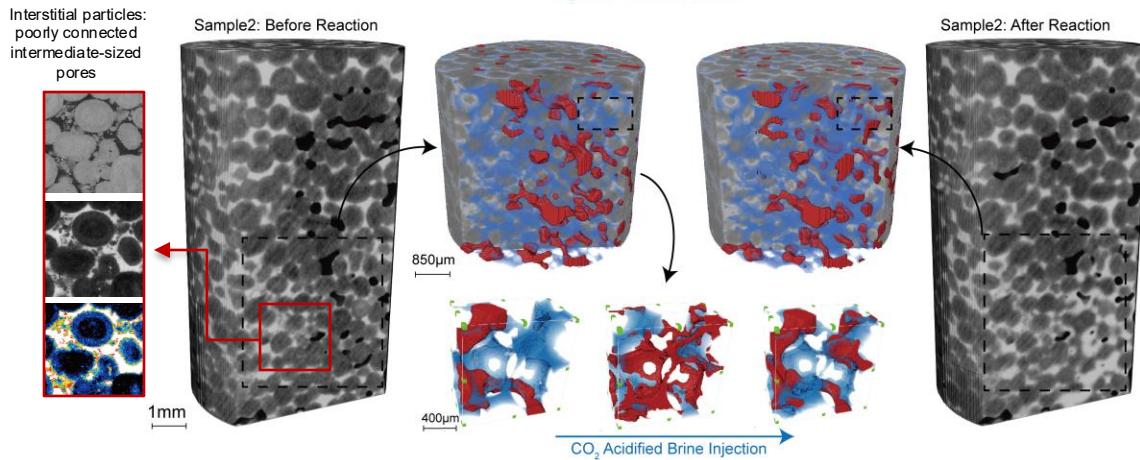


Gray: Rock
White: Brine
Black: Oil



A positive feedback loop:

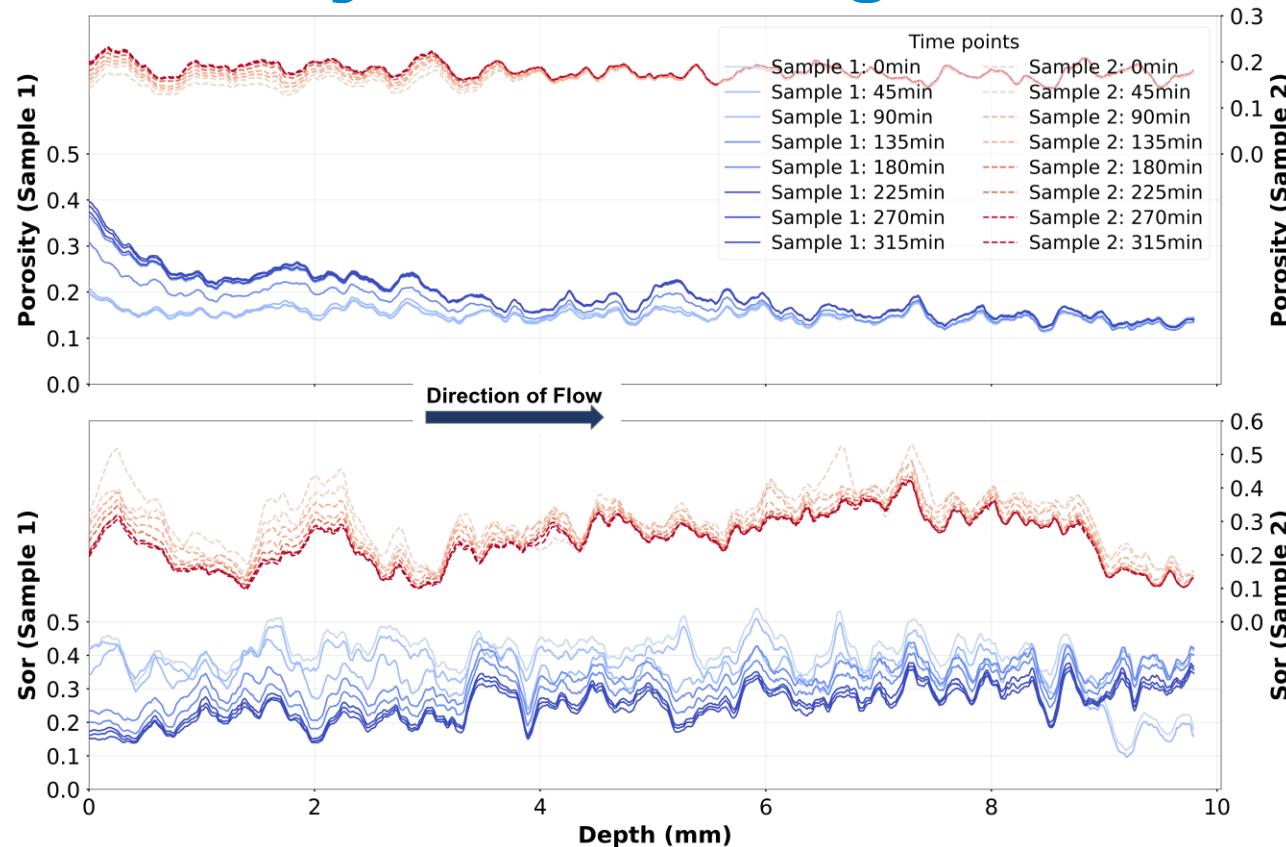
- Displacement-driven channel growth and widening are enhanced by dissolution, which further increases displacement.



A suppressed regime:

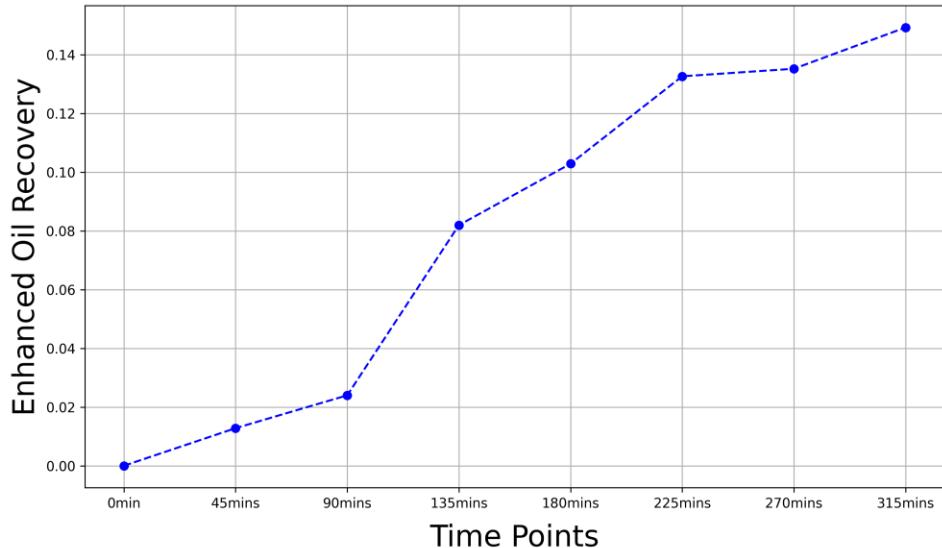
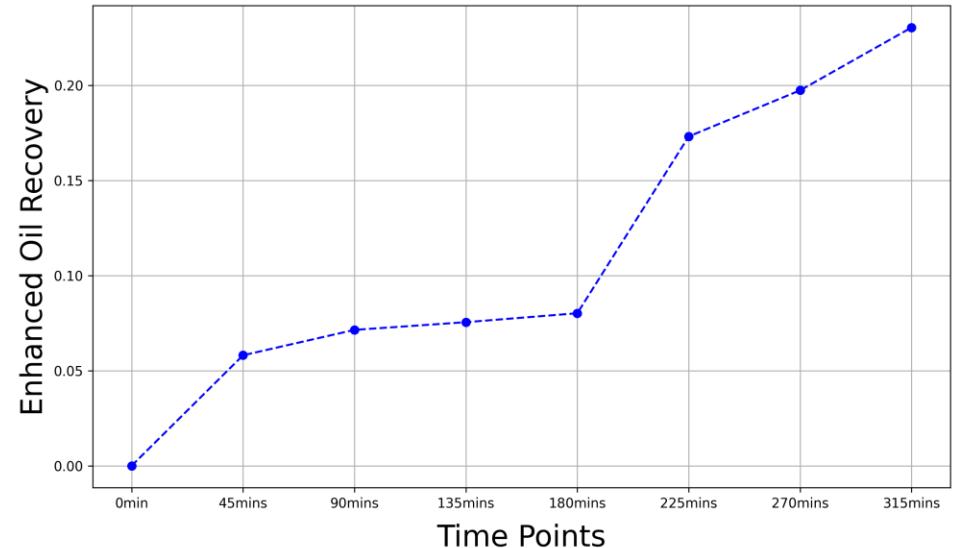
- Fine-scale heterogeneity in low-connectivity regions limits channel formation, while oil blockage prevents dissolution and channel breakthrough.

● Porosity and remaining oil saturation



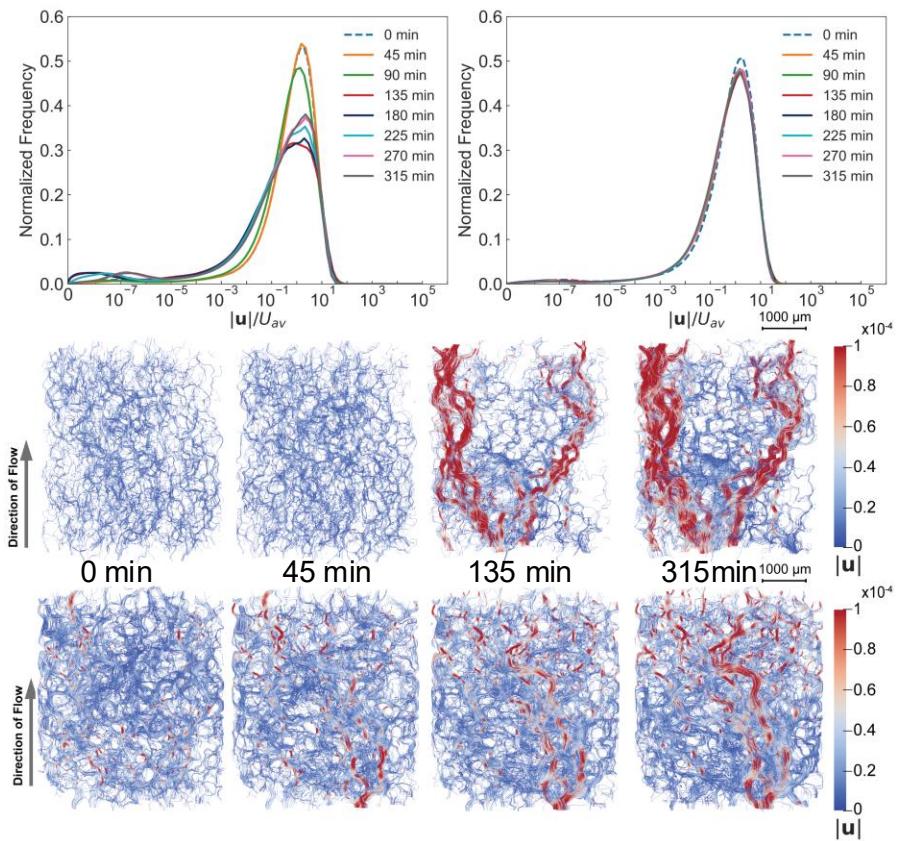
- The changes in Sample 1 are much more significant.
- Sample 2 shows only modest and localized changes at the inlet, confirming a spatially confined reaction.

• Additional oil recovery



- The injection of CO₂ saturated brine facilitates the recovery of residual oil.

- **Flow field evolution**



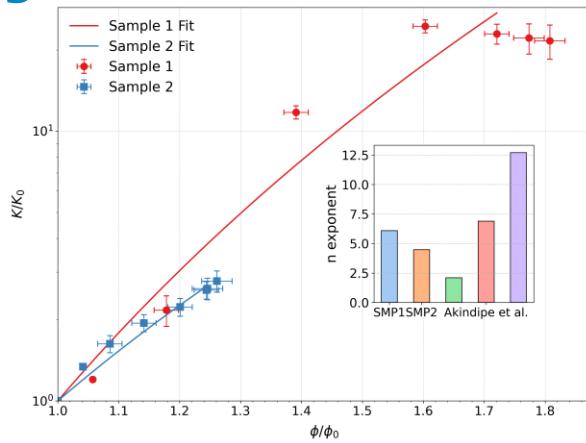
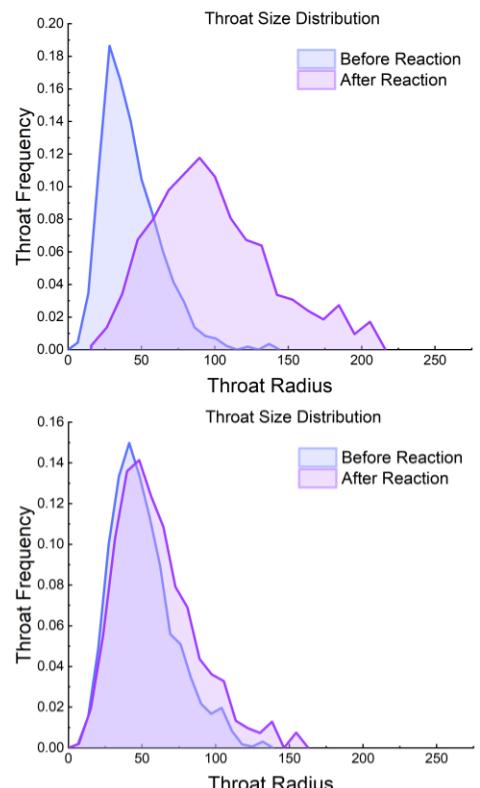
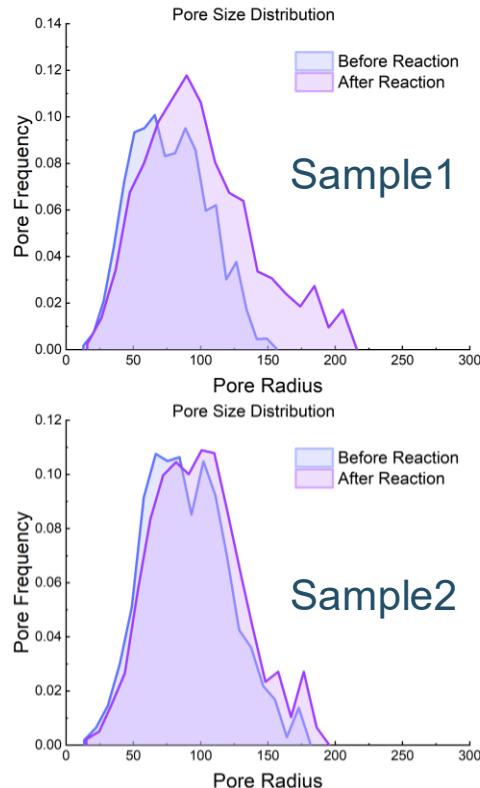
Sample1

- Fast-flow channels emerged, breaking the initial flow uniformity.
- Velocity heterogeneity increased, with more stagnant regions observed.

Sample 2

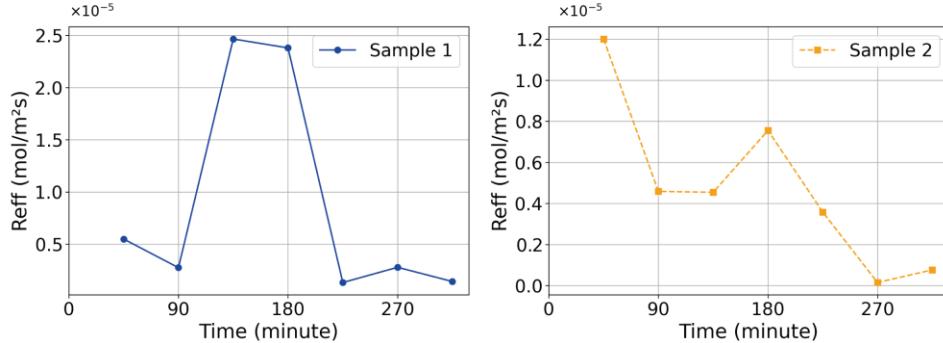
- Bottom-localized channels, with dispersed flow at the top.
- Velocity distribution in Sample 2 changed less over time.

• Pore occupancy and permeability



- The reaction enlarges pores and throats, increasing effective porosity and significantly enhancing permeability by improving the connectivity and fluid flow capacity of the sample.

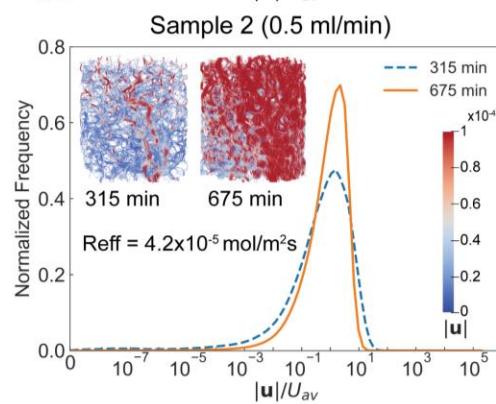
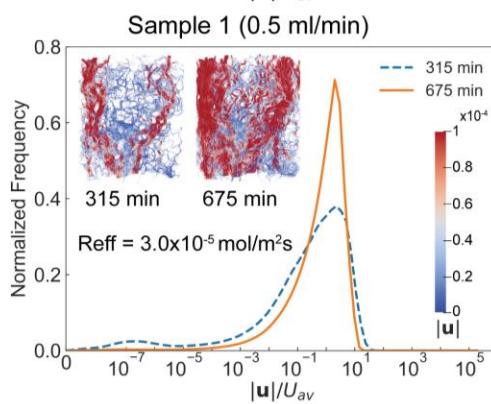
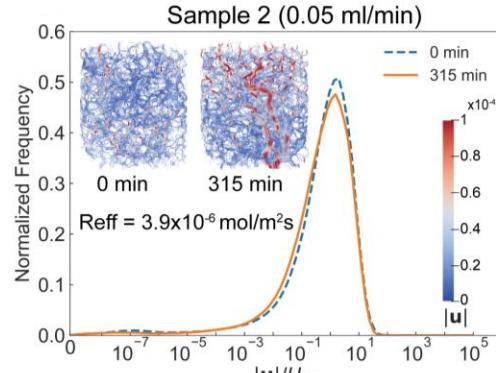
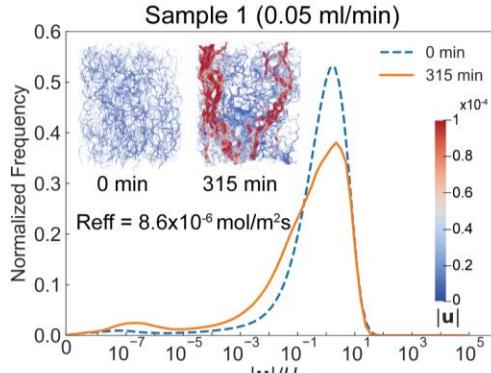
• Effective reaction rate at low flow rate



Condition	Reaction Rate ($\text{mol}/\text{m}^2\text{s}$)	
Ketton limestone multiphase	Sample 1	Sample 2
Low flow rate	8.6×10^{-6}	3.9×10^{-6}
High flow rate	3.0×10^{-5}	4.2×10^{-5}
Silurian dolomite single-phase	Heterogeneity A	Heterogeneity B
Low flow rate	3.15×10^{-6}	1.35×10^{-6}
High flow rate	5.47×10^{-6}	10.6×10^{-6}
Ketton limestone single-phase		5.0×10^{-5}
Batch reaction rate ³⁵		6.9×10^{-4}

- Sample 1: a sharp initial reaction rate increase followed by a decline.
- Sample 2: a stable but lower reaction rate
- Reaction rates in multiphase flow are significantly lower compared to single-phase flow and batch experiments.

• High flowrate vs. Low flow rate



- Sample 1: a sub-linear, three-fold reaction rate increase.
- Sample 2: a near-linear reaction rate increase, surpassing Sample 1.
- Higher injection rates narrow velocity distribution, reduce stagnant voxels, and produce symmetric streamlines.

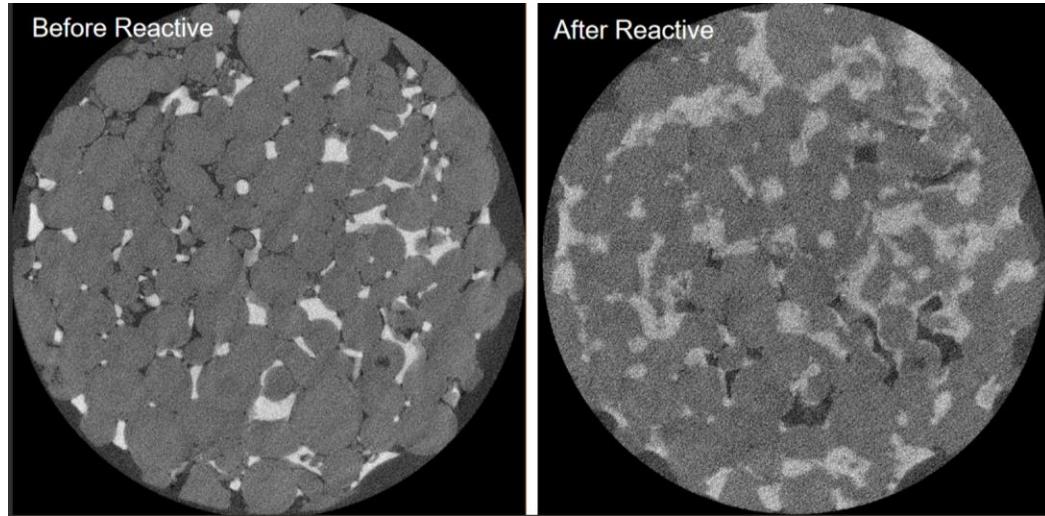
4. Observations and Conclusions

Observations and Conclusions

- Heterogeneity of pore structure, hydrocarbon distribution and remobilization control dissolution patterns and effective reaction rates.
- We capture micron-resolution displacement dynamics, leading to channel formation that enhances CO_2 transport properties.
- Two-phase flow reaction rates are transport-limited and lower than single-phase or batch rates
- Injecting CO_2 -saturated brine facilitates the recovery of residual oil.

5. Future Work

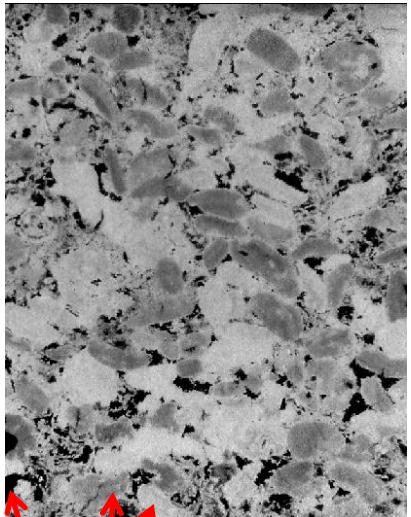
• **Wettability**



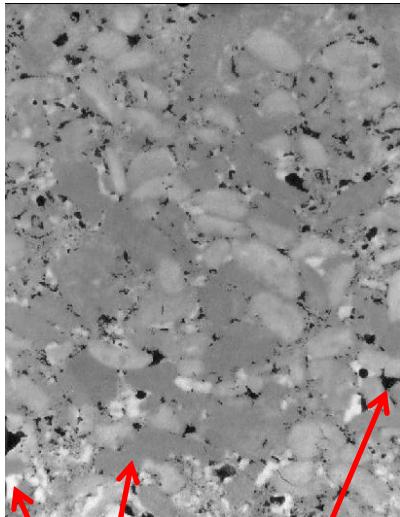
- The reaction rate was noticeably slower compared to the water-wet samples.
- For the first time, we have captured the wettability alteration in rocks caused by CO₂-acidified brine injection.

• Physical heterogeneity

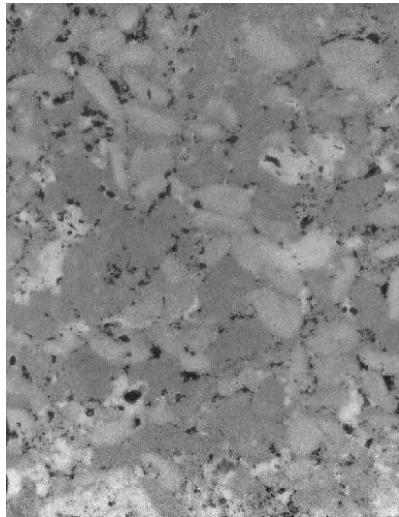
Dry scan



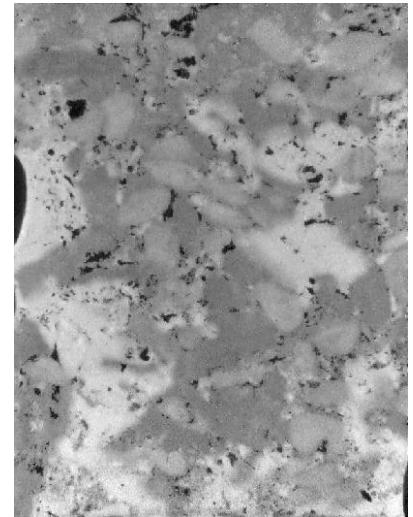
Residual oil saturation scan



Acidified brine injection 10 h



Acidified brine injection 20 h



Gary: Rock
Black: Pore

Gary: Rock
Black: Oil
White: Brine

Acknowledgement

IMPERIAL



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- Lab Staff: Dr. Vincenzo Cunsolo