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# Pore-scale Dynamics of Multiphase Reactive Transport in Carbonates under CO<sub>2</sub>-acidified Brine Injection

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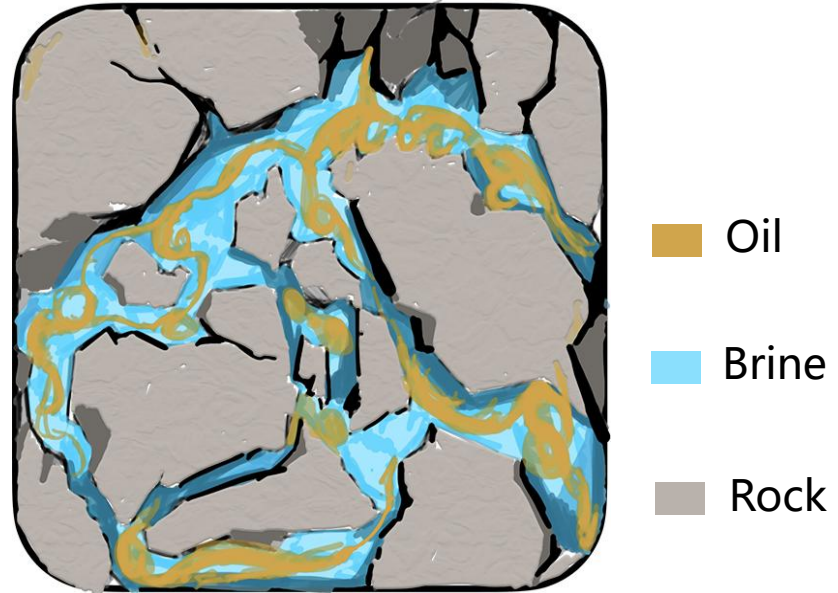
*Qianqian Ma*, Rukuan Chai, Zhuangzhuang Ma, Yanghua Wang, Martin J. Blunt, Branko Bijeljic

*Centre for Reservoir Geophysics · Resource Geophysics Academy · Imperial College London*

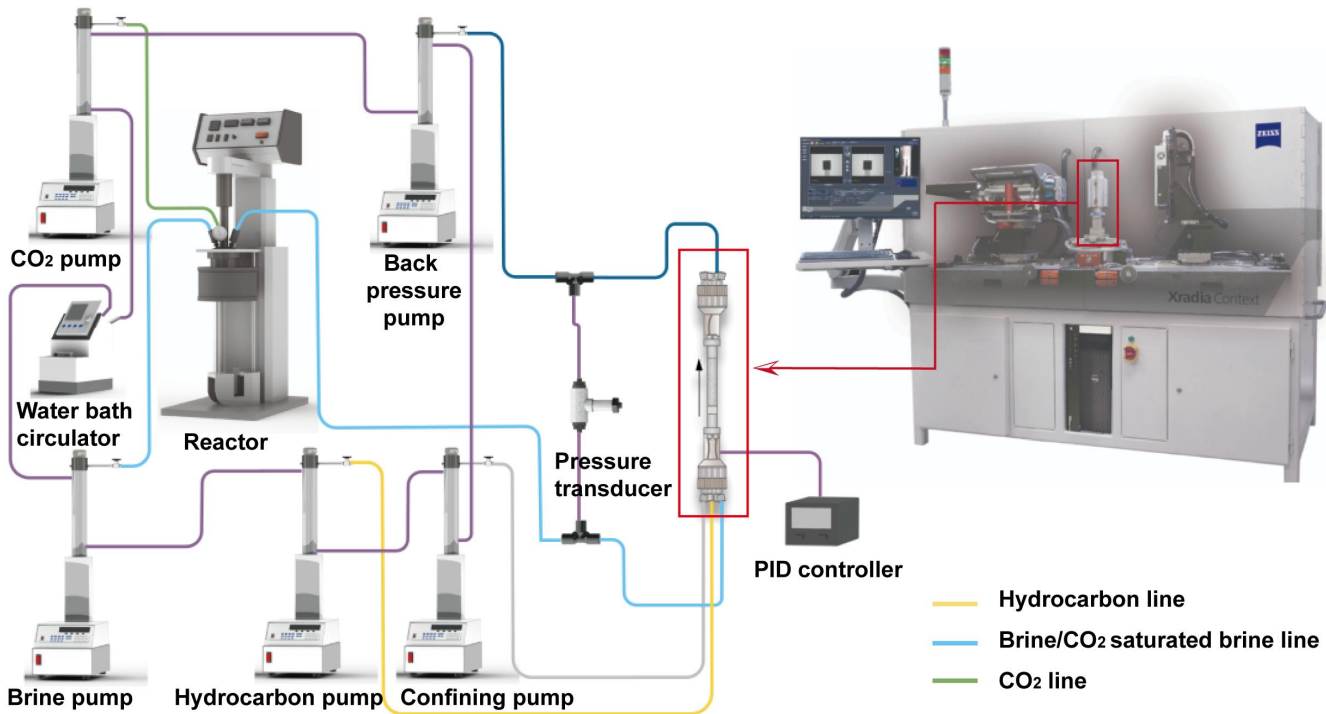
# Challenges

## Reactive transport & multiphase flow

- **Transport heterogeneity:** Hydrocarbon redistributes flow paths across the pore network.
- **Reaction:** Hydrocarbon phase may block the  $\text{CO}_2$  equilibrated brine from reacting.
- **Mass transfer:**  $\text{CO}_2$  partitions into oil ganglia, driving swelling and capillary pressure redistribution.

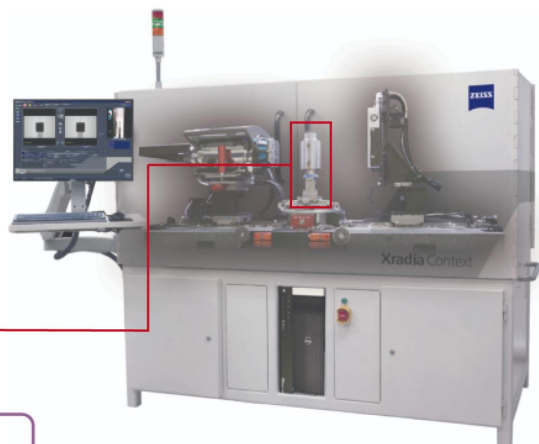


# Experimental apparatus

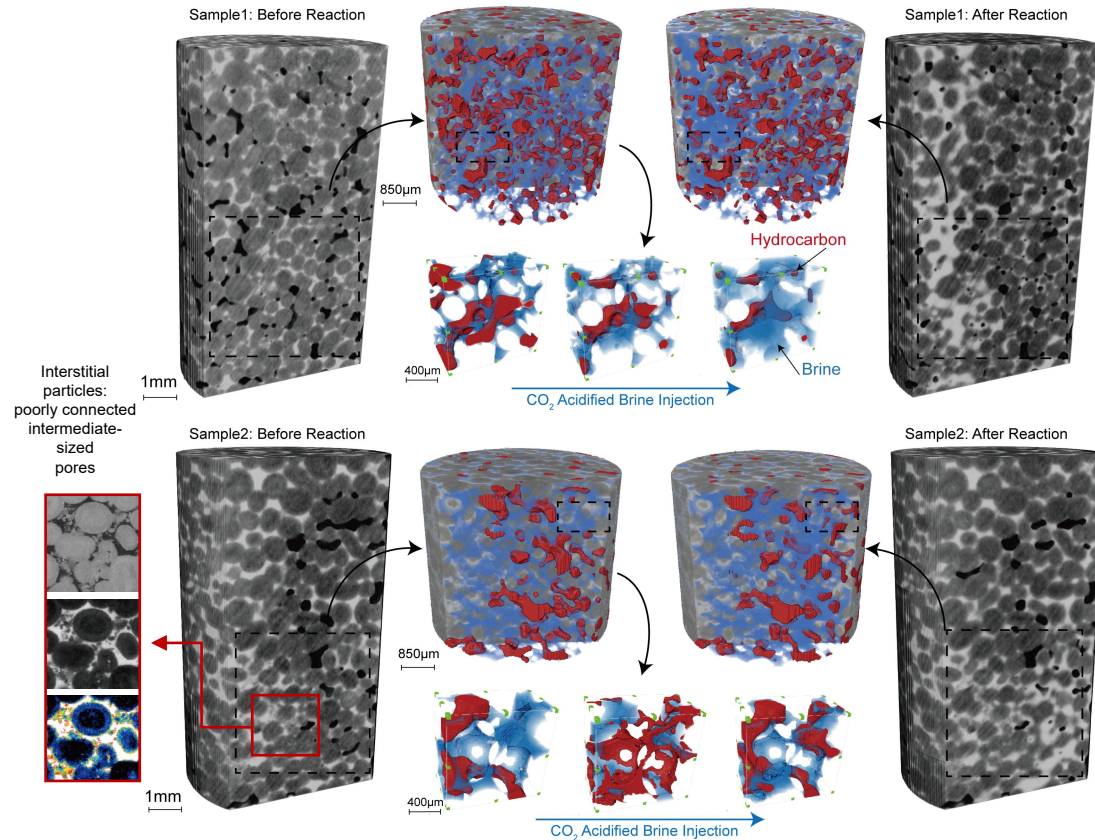


## Measurement System

- 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 Tubing



# Objective 1: How does pore-scale heterogeneity shape dissolution patterns and effective reaction rates?



## ➤ Dual heterogeneity

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

## ➤ Oil mobilisation

- Dissolution enables oil mobilisation; mobilisation reshapes the dissolution pattern.

## ➤ Dynamic evolution

- Structure, fluids, and reaction co-evolve — the dissolution pattern cannot be read from initial conditions.

# Key findings

- Two-phase flow reaction rates are transport-limited and lower than single-phase or batch rates.
- We captured micron-resolution displacement dynamics, leading to channel formation that increased porosity and permeability.
- Heterogeneity of pore structure, hydrocarbon distribution, and re-mobilization controlled the dissolution patterns and effective reaction rates.

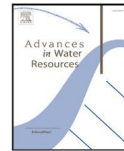
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Pore-scale dynamics of multiphase reactive transport in water-wet carbonates under  $\text{CO}_2$ -acidified brine injection: Dissolution patterns and reaction rates

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Branko Bijeljic <sup>a,\*</sup>



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# Objective 2: How does hydrocarbon swelling and mobilisation compete to govern the effective reaction rate?

Three coexisting phases

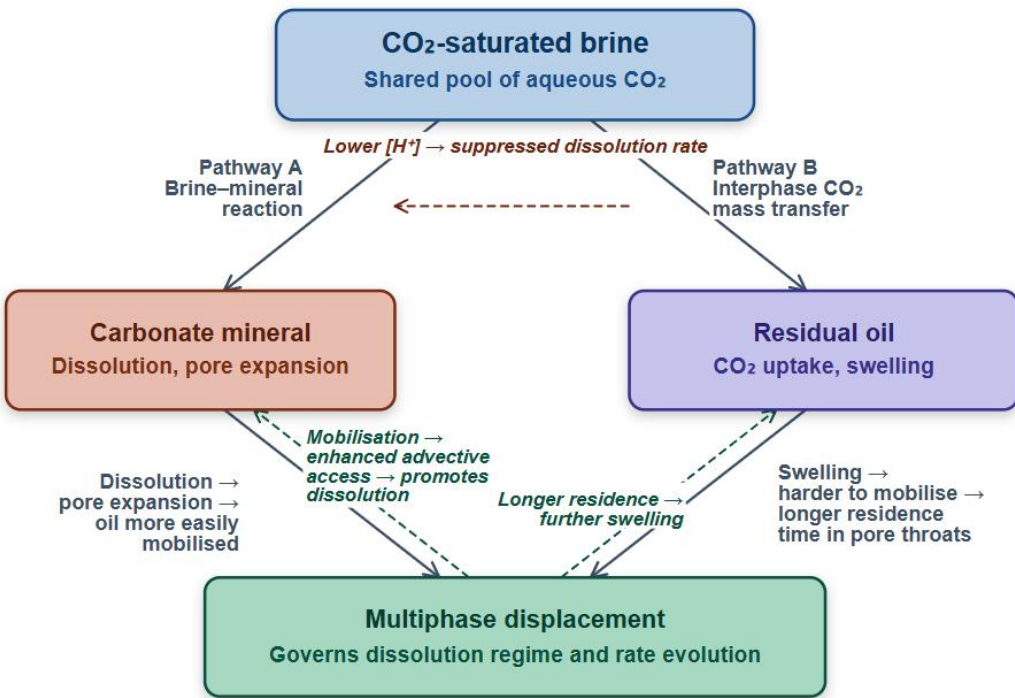
- $\text{CO}_2$ -saturated brine, carbonate mineral, residual hydrocarbon

Two reactive pathways

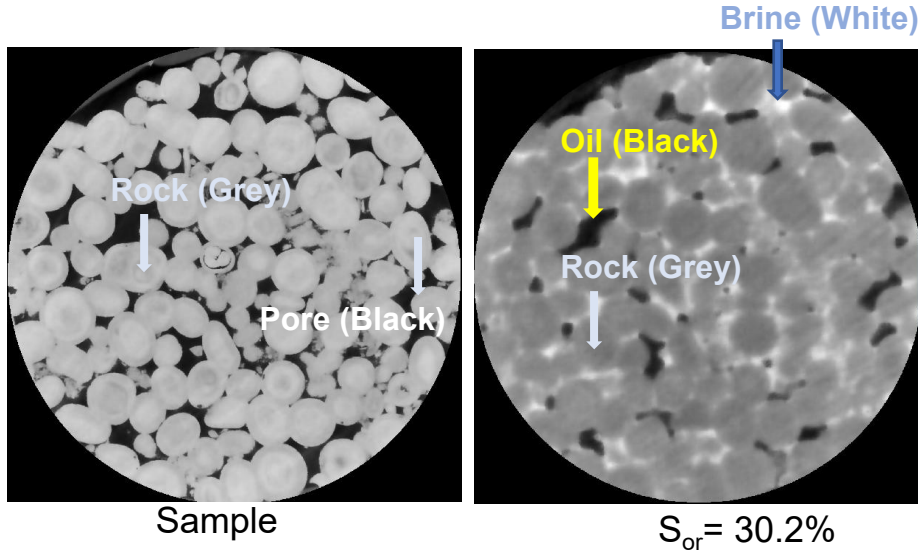
- Brine - mineral reaction  $\rightarrow$  dissolution
- Interphase  $\text{CO}_2$  transfer  $\rightarrow$  hydrocarbon swelling

Coupled through multiphase displacement

- Dissolution enables oil mobilisation
- Swelling prolongs ganglion residence



# Carbonate sample & injection protocol



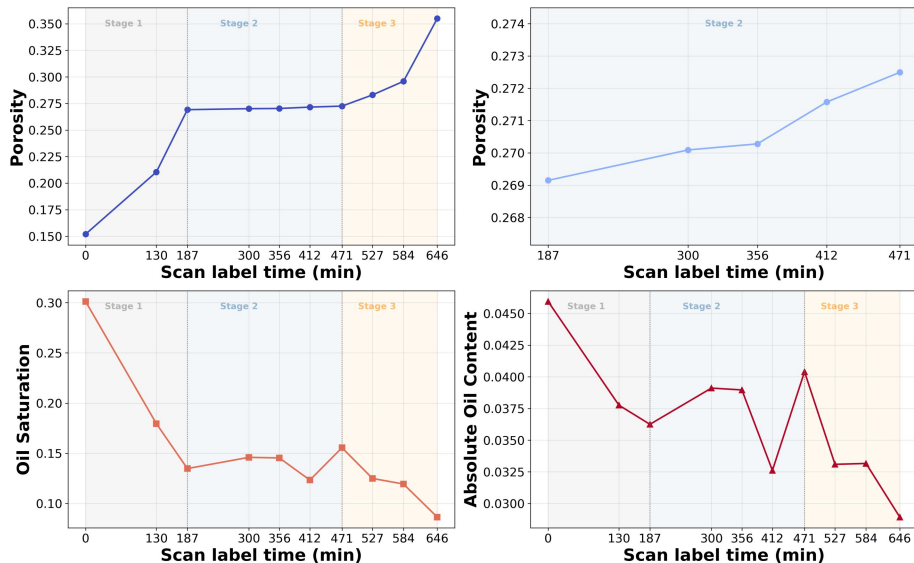
Scan	Time interval (Start–End) (min)	Pore volumes
0 min scan	Residual oil saturation scan	
130 min scan	t=83 to t=130	330
187 min scan	t=140 to t=187	465
300 min scan	t=254 to t=300	677
356 min scan	t=210 to t=356	782
412 min scan	t=366 to t=412	886
471 min scan	t=425 to t=471	990
527 min scan	t=481 to t=527	1094
584 min scan	t=537 to t=584	1195
646 min scan	t=597 to t=646	1294

## Injection Protocol

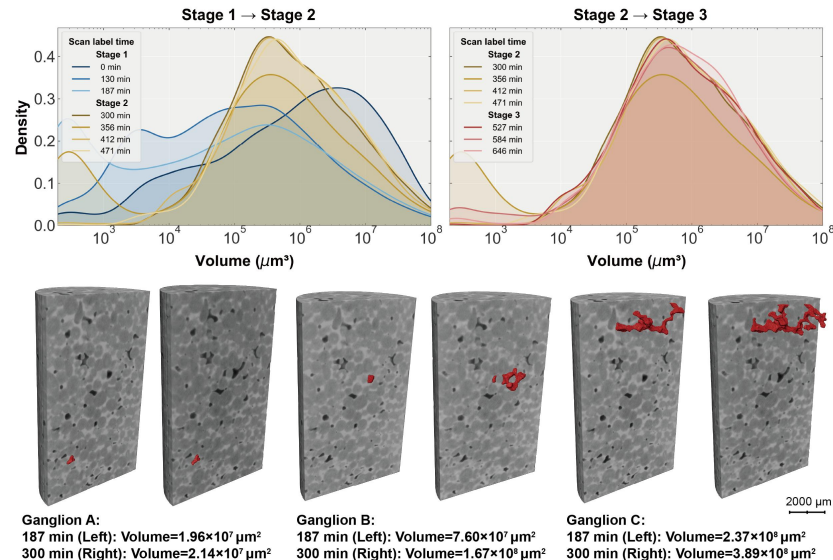
- 0 ~ 73 min: Constant flow at 0.1 mL/min
- 73 ~ 646 min: Cyclic flow — 0.5 mL/min (10 min) → 0.1 mL/min (~ 47 min), × 10
- Total duration: 646 min

# Carbonate dissolution proceeds in three distinct stages

porosity, oil saturation, and oil content

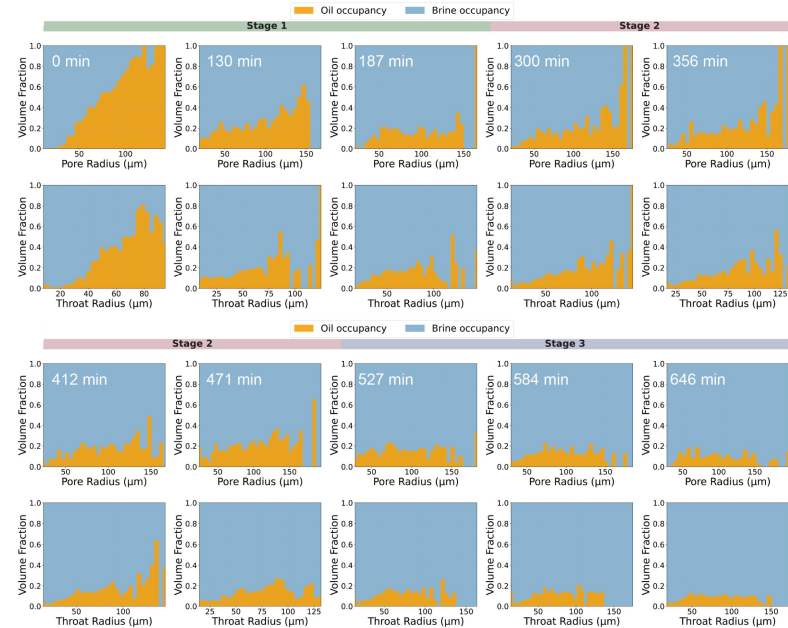
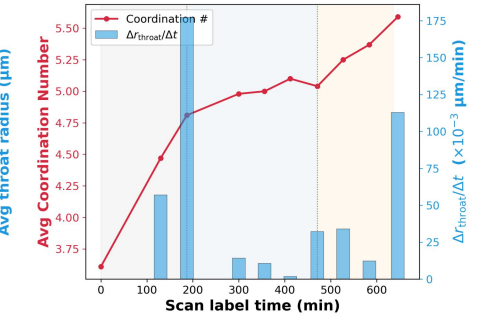
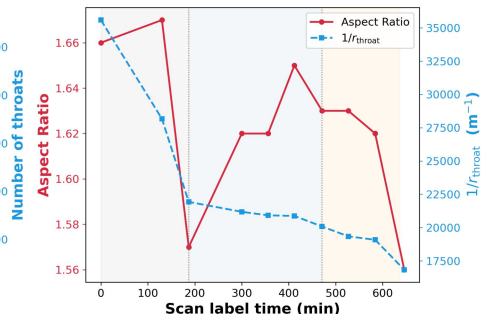
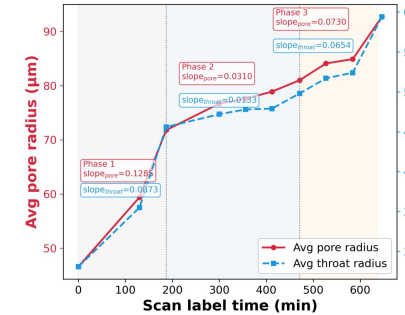
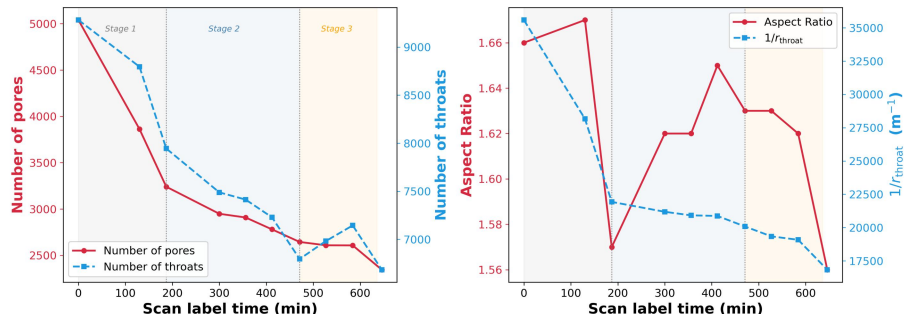


ganglia distribution and size



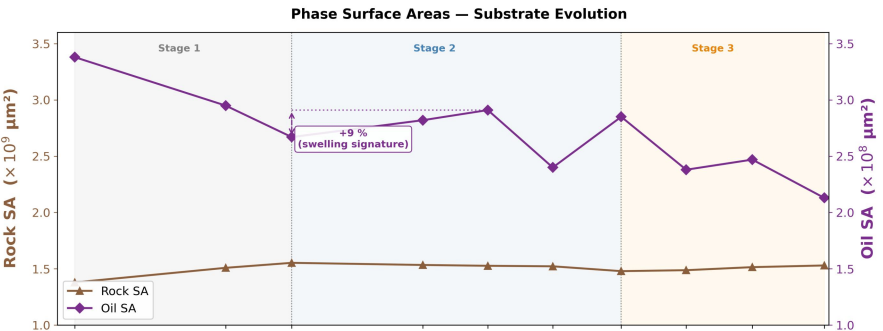
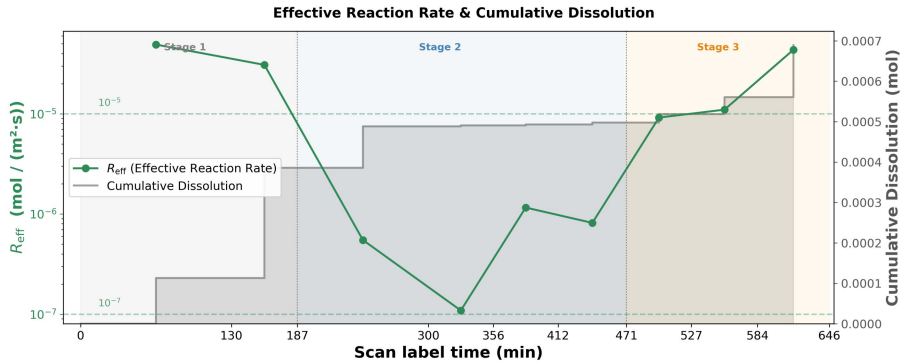
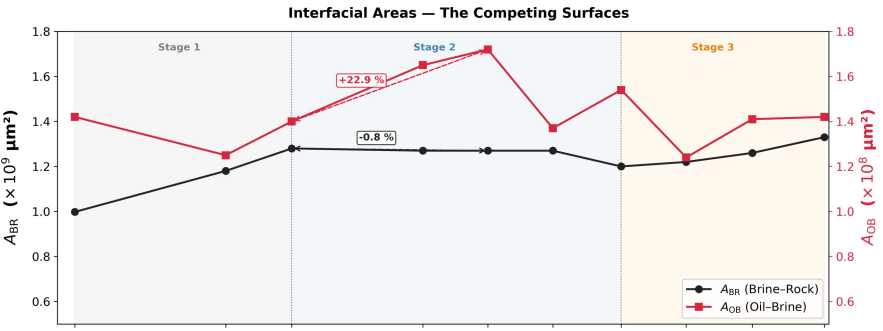
- Stage 1 (0 ~ 187 min): Porosity rises linearly; oil saturation and ganglia size both decrease sharply.
- Stage 2 (187 ~ 471 min): Porosity plateaus; absolute oil volume rebounds — swollen ganglia grow and coalesce in place.
- Stage 3 (471 ~ 646 min): Porosity resumes rising; oil volume declines — under identical injection conditions.

# Pore-space geometry, topology, and fluid occupancy



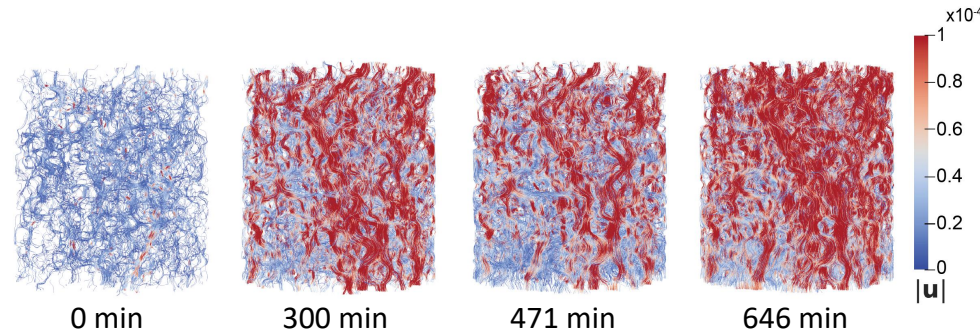
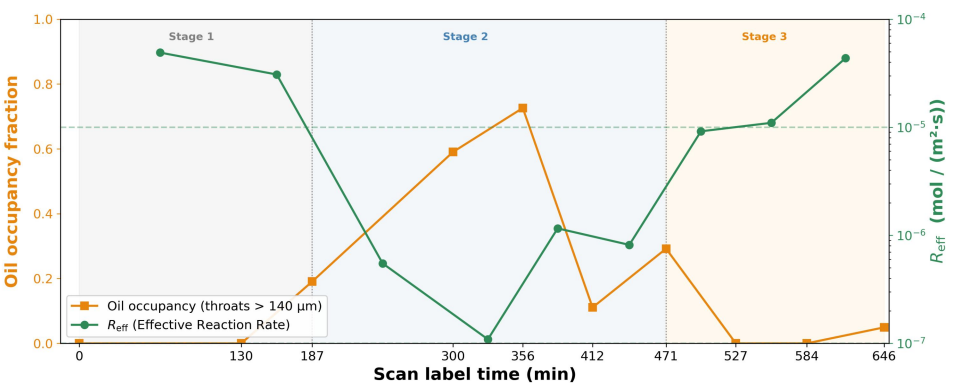
- Stage 1: Dissolution widens throats and enhances connectivity; oil is progressively displaced from the network.
- Stage 2: Swollen ganglia reoccupy the largest throats and block preferential flow paths; occupancy peaks at 73% for throats >140  $\mu\text{m}$ .
- Stage 3: Dissolution reopens flow paths and restores connectivity; oil is rapidly displaced from the largest throats.

# Interfacial area and dissolution rate



- During Stage 2, oil–brine interface grows 23%.
- Brine–rock interface stays flat.
- The effective reaction rate is non-monotonic over time and decreases a hundredfold. Much lower than batch (uniform) reaction rate.

# Why does carbonate dissolution nearly stop and then recover?



## Geometric factors dominate over chemical

- Chemical — ganglia absorb  $CO_2$  and reduce  $H^+$  availability in the bulk brine.
- Geometric — swollen ganglia blocks acidic brine access to solid surface.

## Mobilisation drives reaction rate recovery

- Once dissolution opens bypass routes, ganglia are remobilised, advective contact is restored, and the reaction rate recovers.

## Competition:

- Swelling vs mobilisation governs advective access.

# Conclusions

- Time-resolved micro-CT captures three stages of carbonate dissolution during CO<sub>2</sub>-saturated brine injection in hydrocarbon-bearing rock.
- Dissolution is initially rapid and then drops a hundredfold as hydrocarbon swells and blocks access of acidic brine to the mineral surface.
- The effective dissolution rate reflects a competition between hydrocarbon swelling and ganglion mobilisation, which together govern advective access to mineral surfaces and cause non-monotonic reaction behaviour over time.
- Multiphase reactive transport introduces additional dynamic complexity, which should be considered in the design of CO<sub>2</sub> storage

# Thank You

*Questions & Discussion*

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### Publications

**Paper 1:** Ma et al., *Advances in Water Resources*, 208 (2026), 105202

**Paper 2:** arXiv preprint



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