IMPERIAL

Pore-scale Reactive Transport Mechanisms of Carbon-Dioxide Storage in Depleted Hydrocarbon Fields

Qianqian Ma

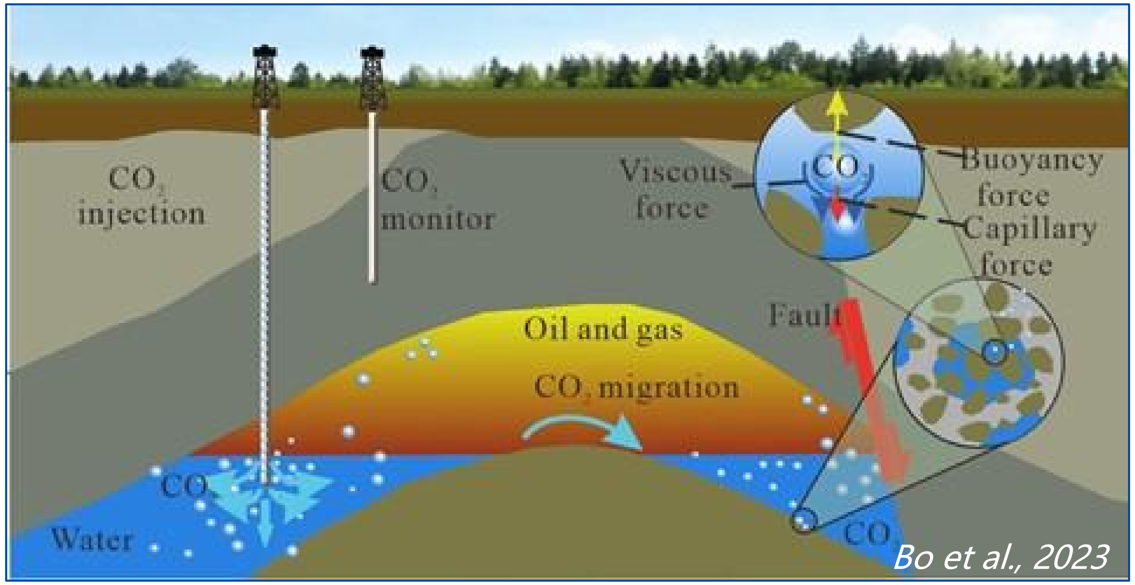
Supervised by: Dr. Branko Bijeljic; Prof. Martin Blunt

2025-01-15

IMPERIAL

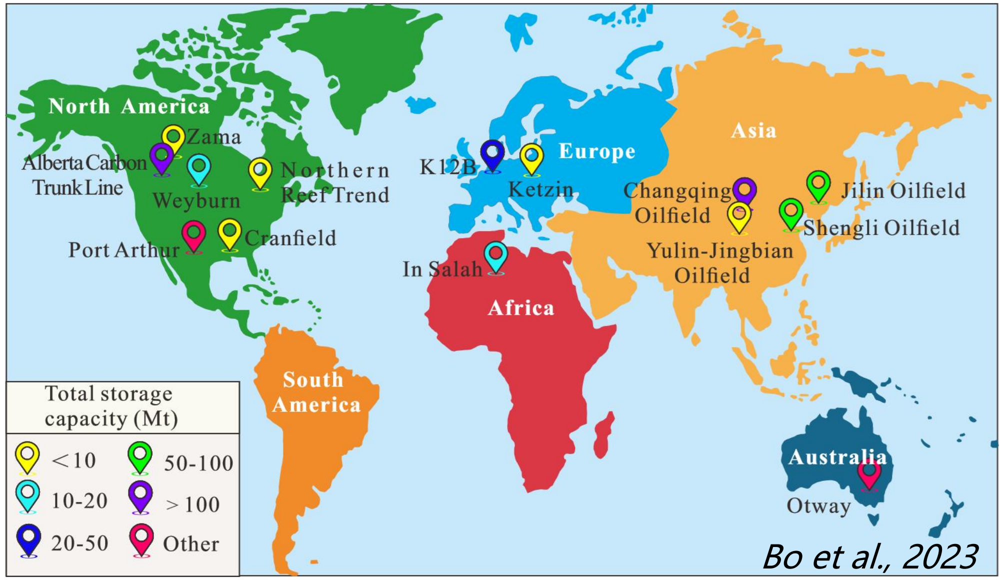
CONTENTS

• Research Background



Depleted Hydrocarbon Fields

Less additional costs to implement
Mature technical processes
Ease of management



Global CO_2 Storage Projects in Depleted Hydrocarbon Fields

Global CO_2 storage capacity of 900-1200Gt

➢ Depleted hydrocarbon fields have obvious advantages compared to other storage methods in terms of CO_2 storage capacity, reservoir characterization experience, existing oil and gas well infrastructure, and operability of storage[1] .

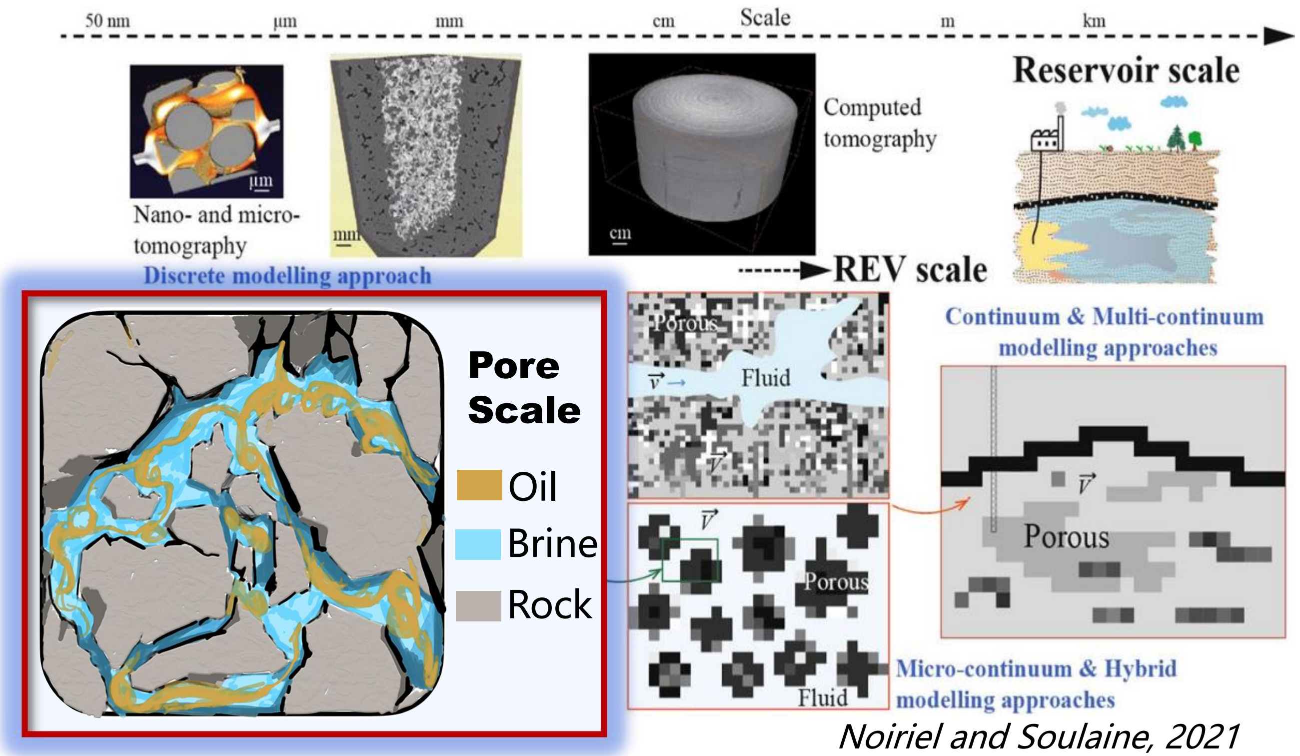
Pore-scale study of reactive transport in the presence of oil

Pore-scale

- Pore-scale processes
- Reactive transport phenomena

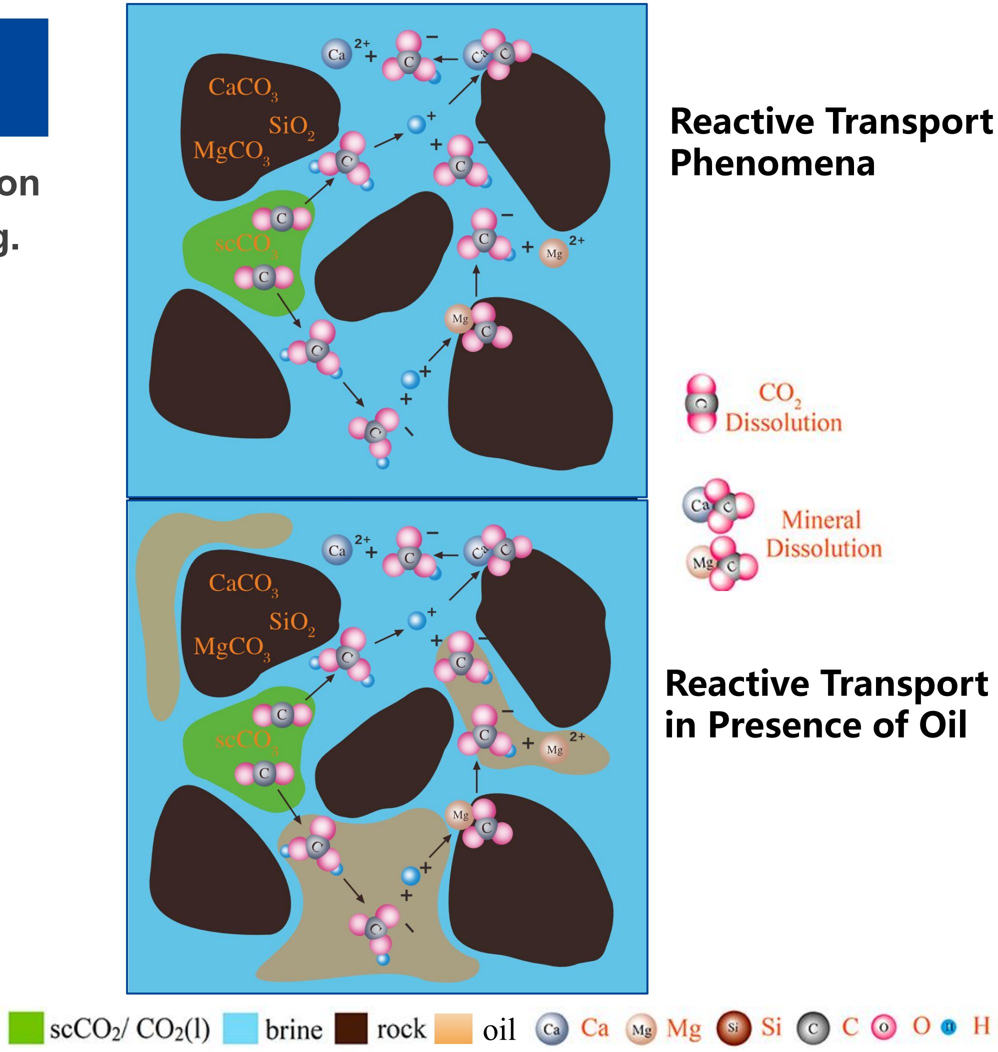
Continuum-scale

- Homogeneous and isotropic region
- Phenomenological parameters eg. permeability etc



Noiriel and Soulaine, 2021

Integration of approaches for reactive transport from pore to reservoir scale[2]



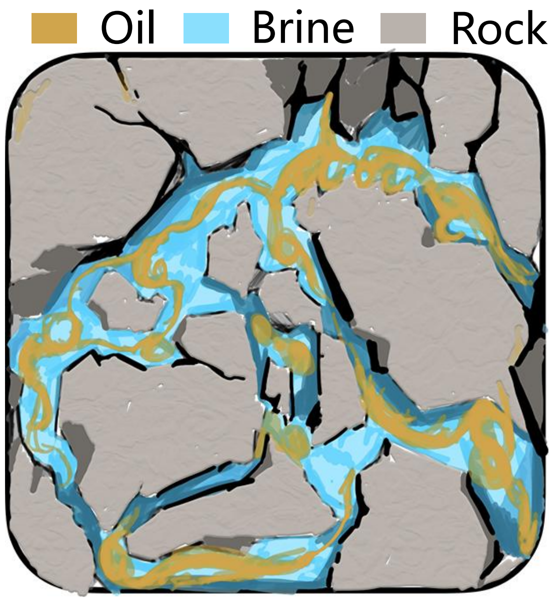
Reactive transport phenomena[3] *Xu et al., 2017*

Challenges

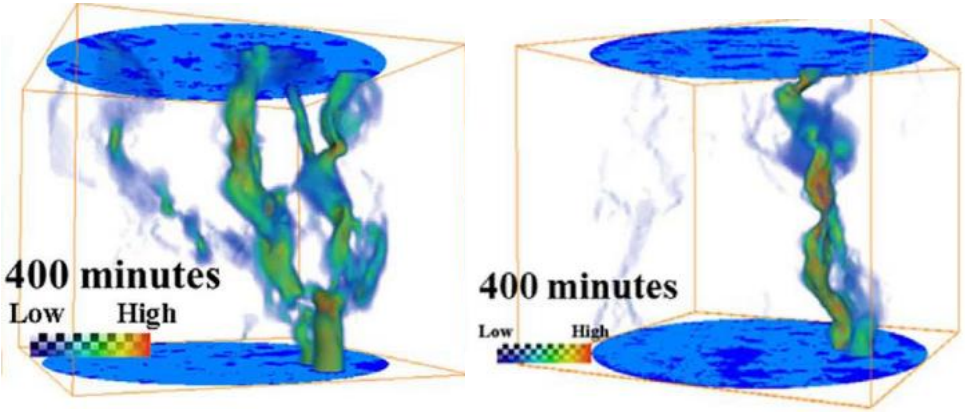
Reactive Transport & Multiphase flow
Carbonate rock partially saturated with hydrocarbon phase.

Heterogeneity
Hydrocarbon phase will change the transport heterogeneity and may block the CO_2 equilibrated brine from reacting.

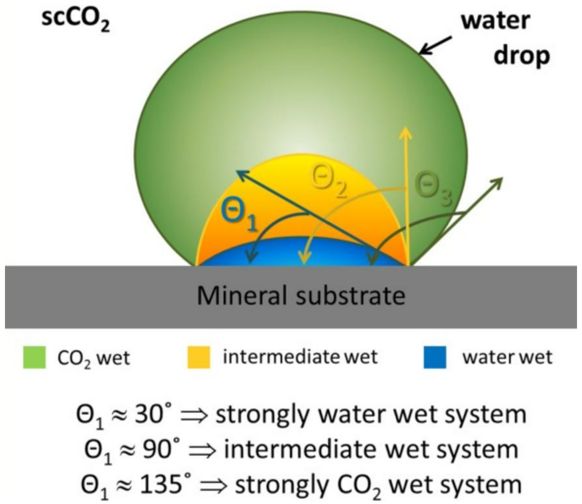
Wettability
Mineralogical complexity and heterogeneity.



Carbonate rock saturated with oil and brine



Al-Khulaifi et al., 2017
Velocity fields showing changes with dissolution for the sample with Heterogeneity A & B[4]



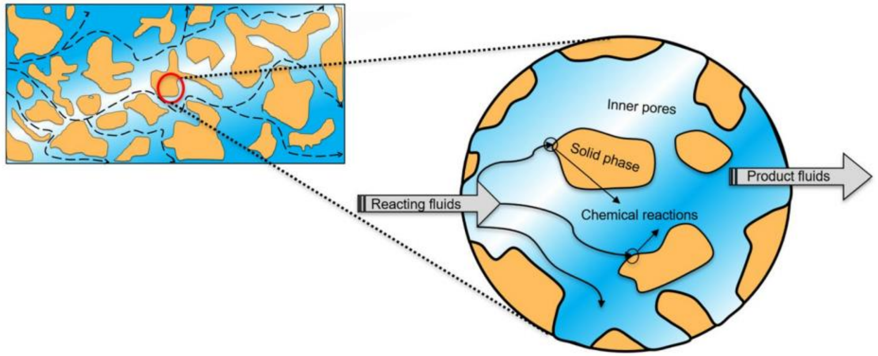
Wang et al., 2024
Wettaility States[5]

• Research Objectives

1. To devise an X-ray imaging and image-based modelling methodology to study reactive transport during CO_2-acidified brine injection into hydrocarbon reservoirs.

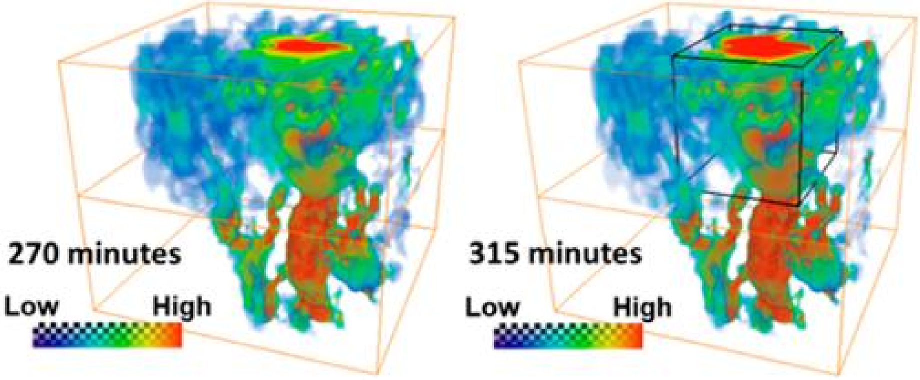
2. To study the impact of transport heterogeneity on flow mechanisms, effective reaction rates and dissolution patterns in a Ketton limestone partially saturated with hydrocarbon phase under two remaining oil saturations.

3. To study the impact of wettability on effective reaction rates and dissolution patterns under two different wettability states.



Abd and Abushaikha, 2021

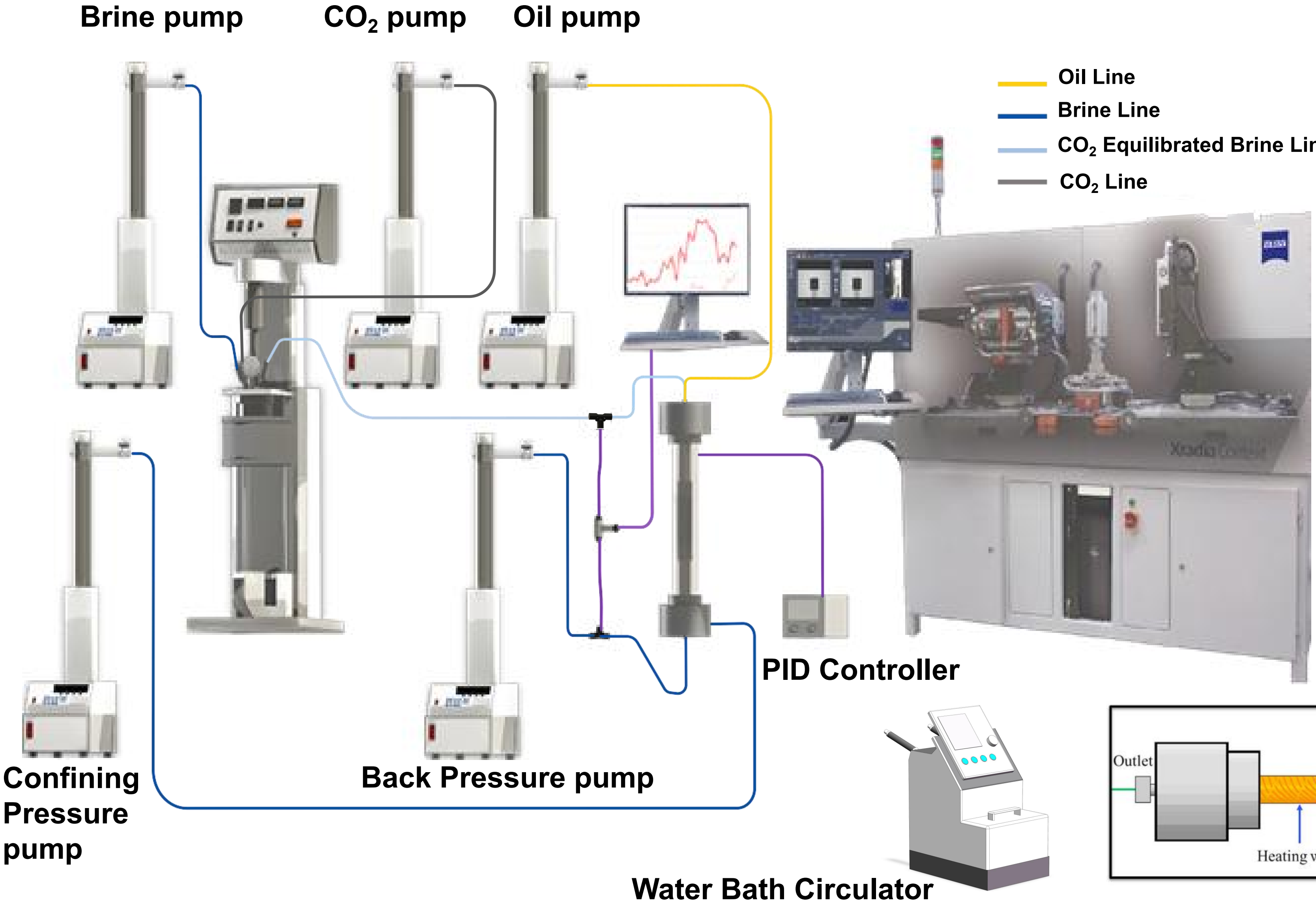
The reaction sites of the flowing fluids with the ions present in the porous media[6]



Menke et al., 2016

3D renderings of flow velocities from modeling over the span of the experiment as dissolution advance[7]

Experimental apparatus



Measurement System

1. Fluid Flow Unit

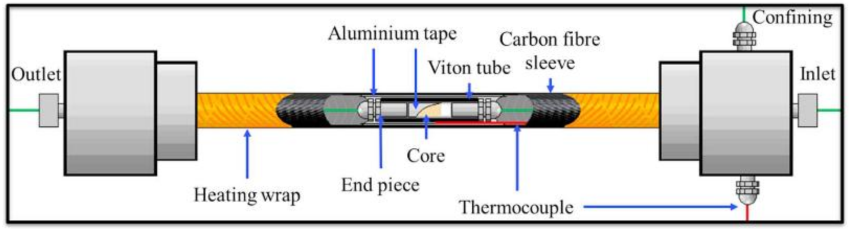
ISCO Pumps
High T&P Reactor
High T&P Core Holder
Back Pressure Regulator

2. Monitoring Unit

X-ray CT
Pressure Transducer

3. Stability Unit

Water Bath Circulator
PID Controller
Insulation Cover for Tubings



(Al-Khulaifi et al., 2017)

Experimental procedure

Preparation Stage → Pre-Reaction Stage → Reaction Stage → Post-Reaction Stage

Experimental procedure: Preparation Stage

Preparation Stage

- Carbonate Sample Preparation
- Porosity & Permeability Measurement
- Pre-reaction SEM Imaging

Experimental procedure: Pre-Reaction Stage

Preparation Stage → **Pre-Reaction Stage**

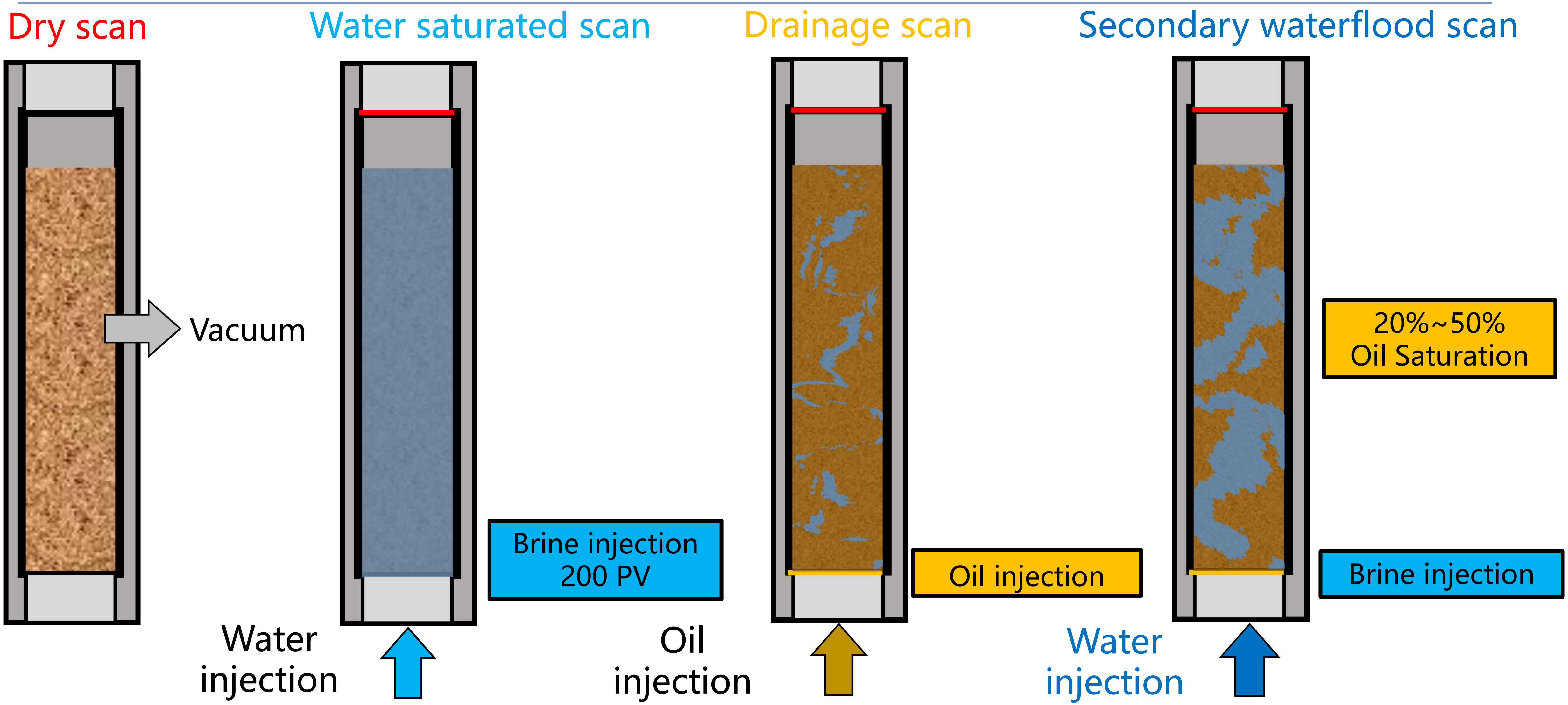
Preparation Stage:
- Carbonate Sample Preparation
- Porosity & Permeability Measurement
- Pre-reaction SEM Imaging

Pre-Reaction Stage:
- Dry Scan
- Water Saturated Scan
- Drainage Scan
- Imbibition Scan

OBJECTIVE 2: Transport Heterogeneity
- Water-wet Ketton limestone will be saturated with oil and brine in different saturation.
- Approximately 20% Sor & 50% Sor
- Quantitatively characterize transport heterogeneity based on their velocity distribution.

OBJECTIVE 3: Wettability
- Samples with different wettability and similar Sor.
- Water-Wet & Oil-Wet.

• Experimental procedure: Pre-Reaction Stage



Pre-Reaction Process
(Original image by Anin Patmonoaji. Modifications made by the Auther.)

Experimental procedure: Reaction-Stage

Preparation Stage → Pre-Reaction Stage → Reaction Stage

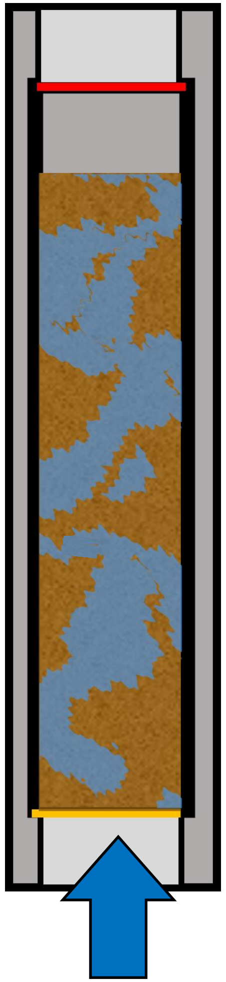
Preparation Stage
- Carbonate Sample Preparation
- Porosity & Permeability Measurement
- Pre-reaction SEM Imaging

Pre-Reaction Stage
- Dry Scan
- Water Saturated Scan
- Drainage Scan
- Imbibition Scan

Reaction Stage
- Reactive Brine Injection
- Dynamic Scanning
- Effluent Sampling

• Experimental procedure: Reaction Stage

Remaining Oil Saturation Reached



Backpressure
8 MPa

Confining
pressure
10 MPa

Scan with X-ray every
half hour.

CO_2 equilibrated brine
injection

Reaction Process
(Original image by Anin Patmonoaji. Modifications made by the Author.)

Experimental procedure: Post-Reaction Stage

Preparation Stage → Pre-Reaction Stage → Reaction Stage → Post-Reaction Stage

Preparation Stage
- Carbonate Sample Preparation
- Porosity & Permeability Measurement
- Pre-reaction SEM Imaging

Pre-Reaction Stage
- Dry Scan
- Water Saturated Scan
- Drainage Scan
- Imbibition Scan

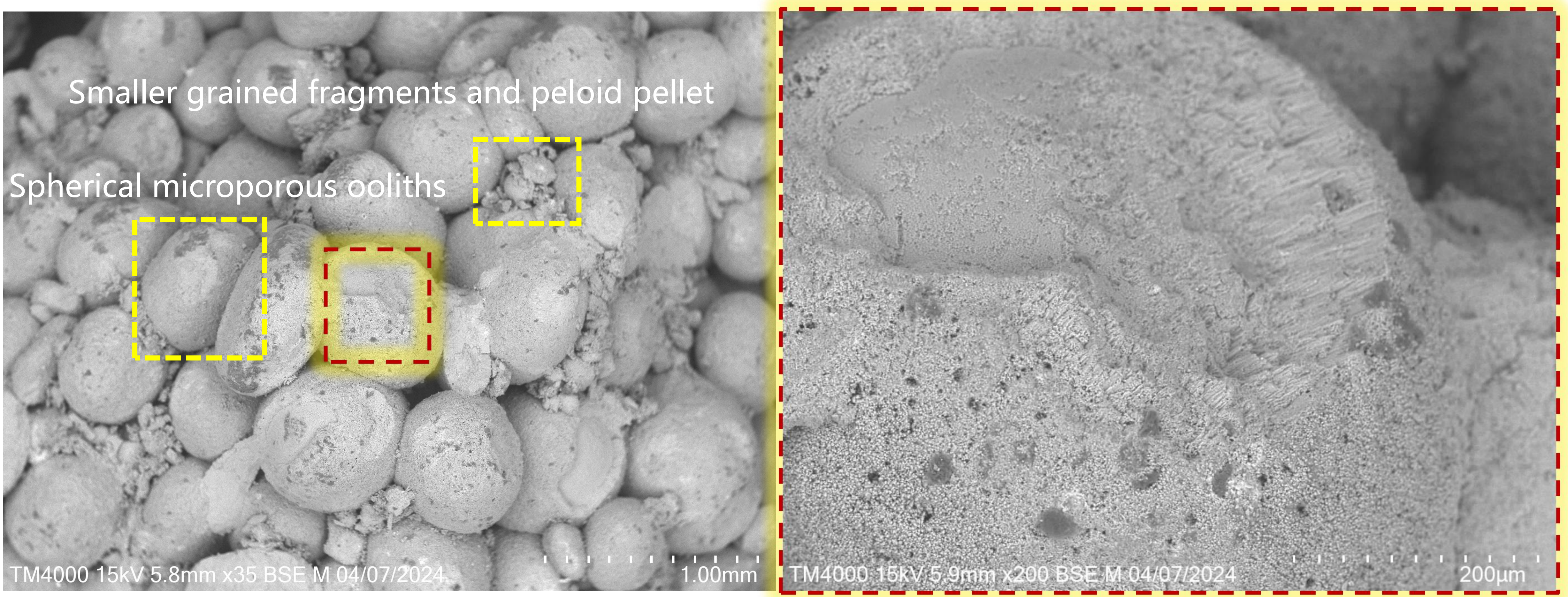
Reaction Stage
- Reactive Brine Injection
- Dynamic Scanning
- Effluent Sampling

Post-Reaction Stage
- Experiment Depressurization
- Core Unloading
- ICP-MS Analysis

• Image Analysis

IMPERIAL

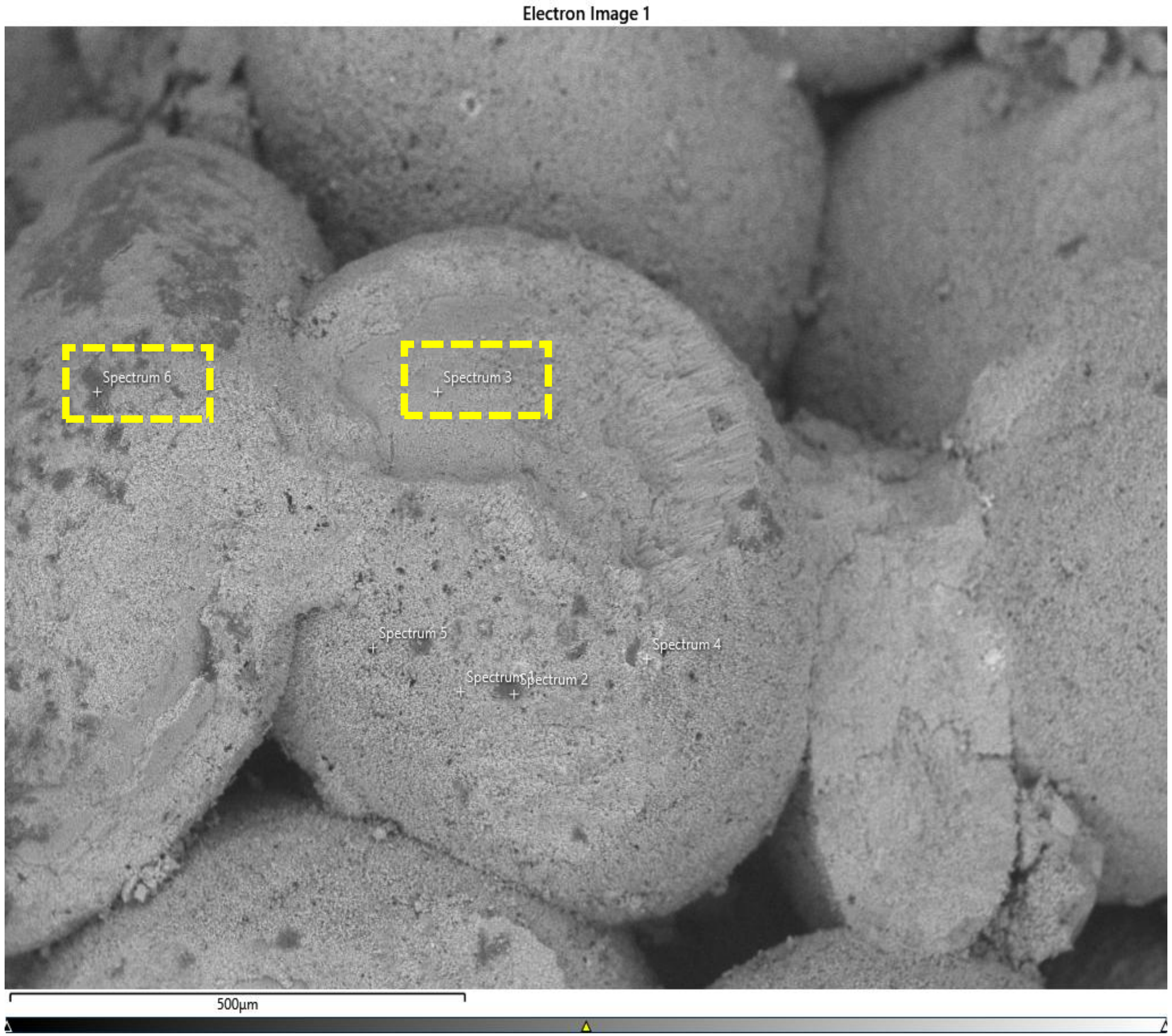
● SEM Analysis

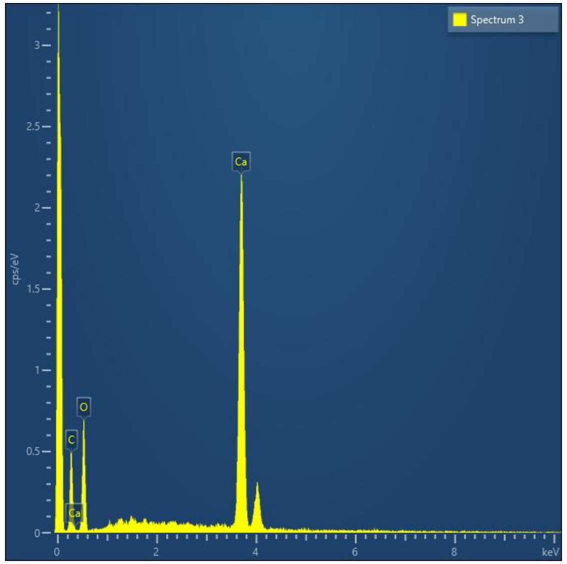


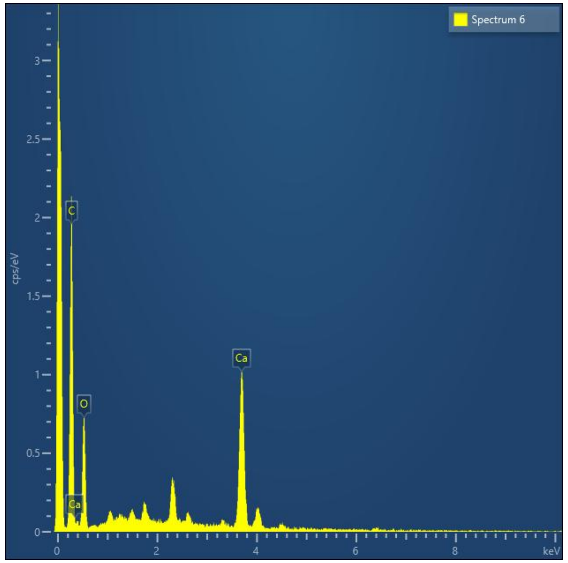
SEM imaging

➢ Physically homogeneous in pore space and grains distribution compared to many other carbonates

SEM Analysis





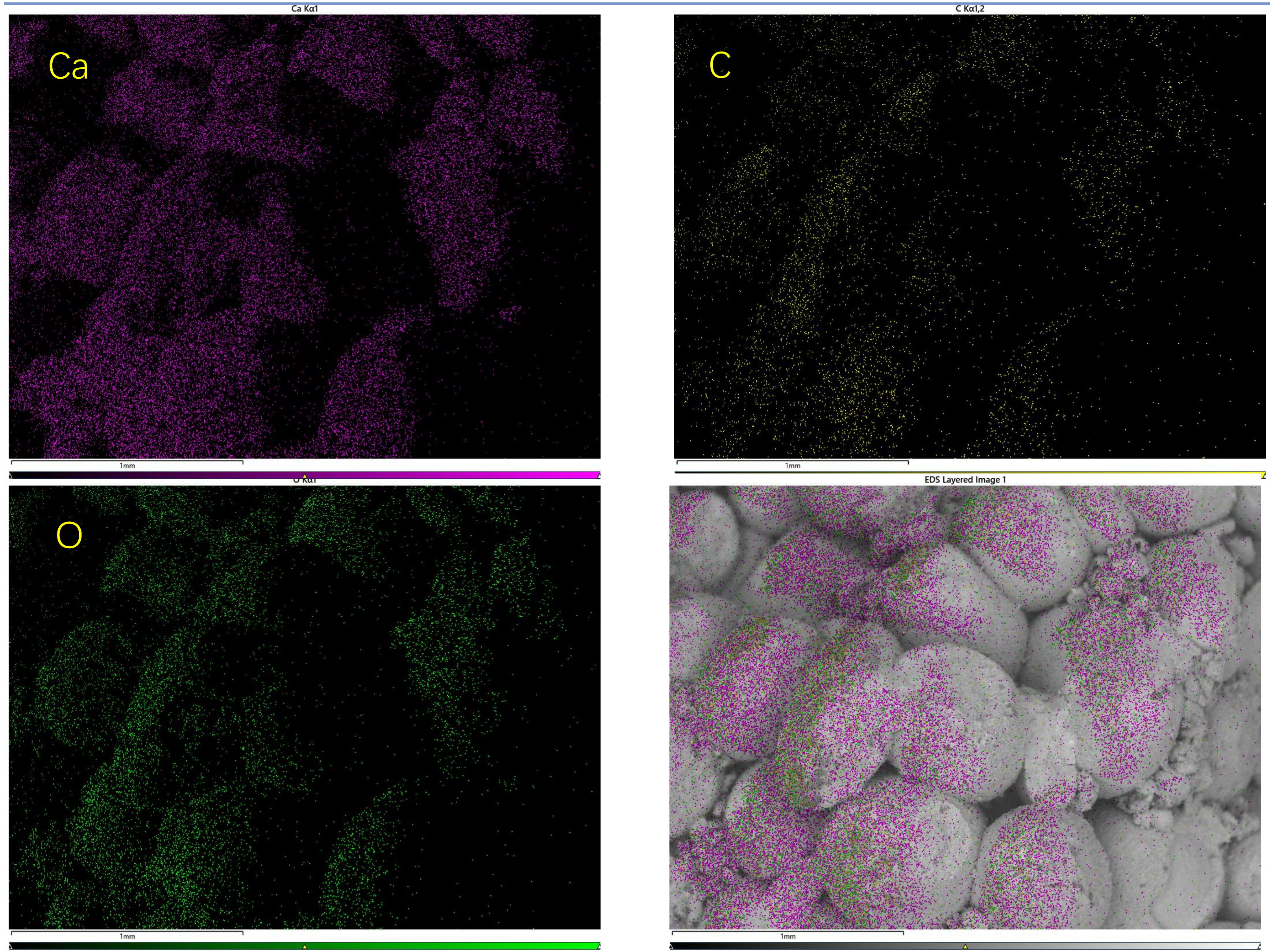


Main Elements:
Ca C O

$CaCO_3$

Element Content

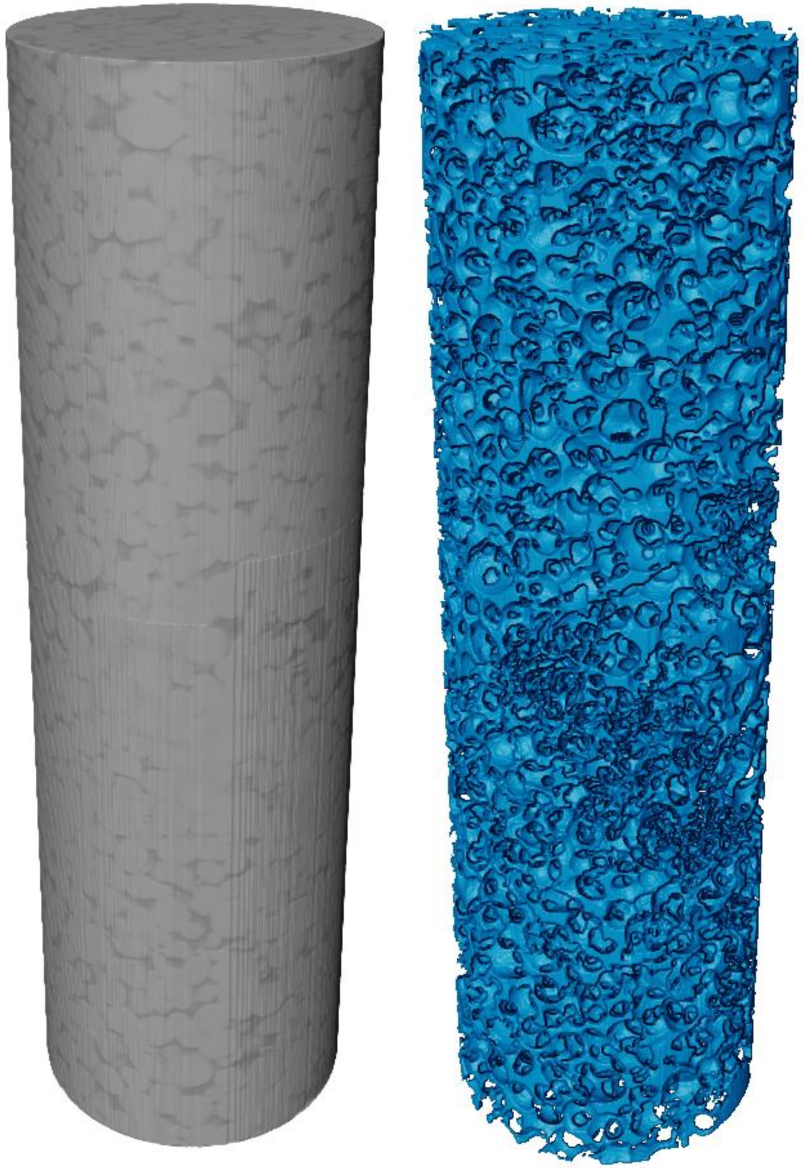
• SEM Analysis



- Chemically homogeneous

Element distribution

• Why Ketton?

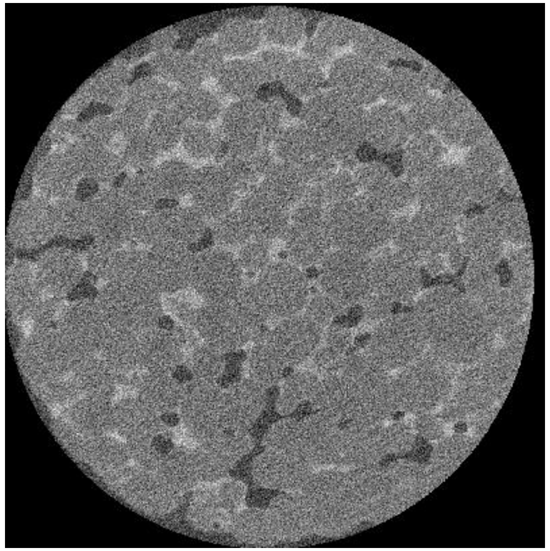


- Ketton is relatively homogeneous in both its **mineralogy and pore-structure**.
- Ketton is a good sample to study the impact of **transport heterogeneity.**
- **Transport heterogeneity will mostly be affected by the introduction of oil.**

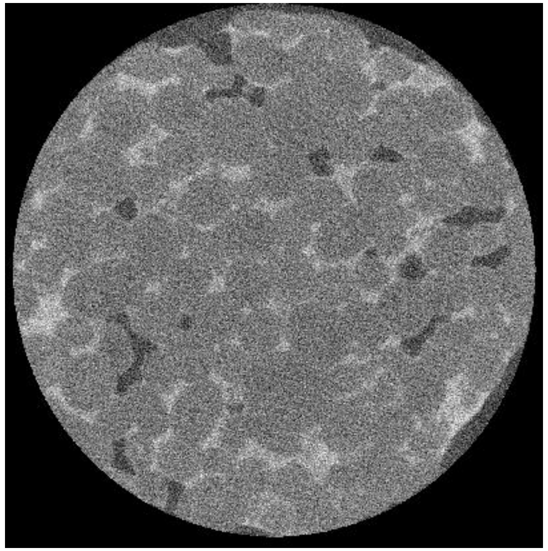
• Experiment Progress

➢ Current Progress: successfully completed the experiments as planned

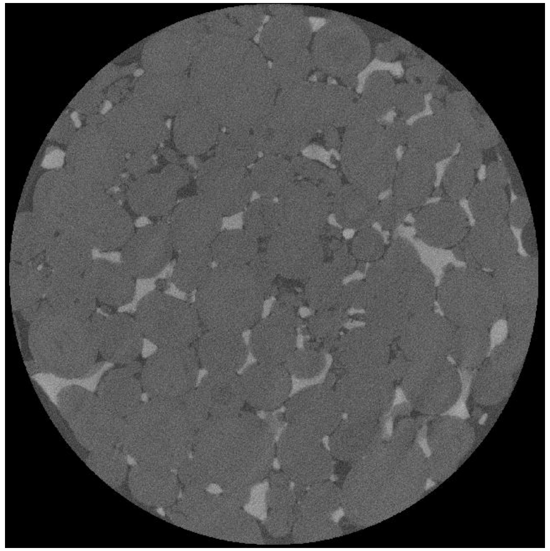
1. **One set of contrast experiments to select parameters for optimal imaging.**
2. **Water-wet samples with different remaining oil saturation.**
3. **One set of oil-wet samples.**



Water-Wet 1#

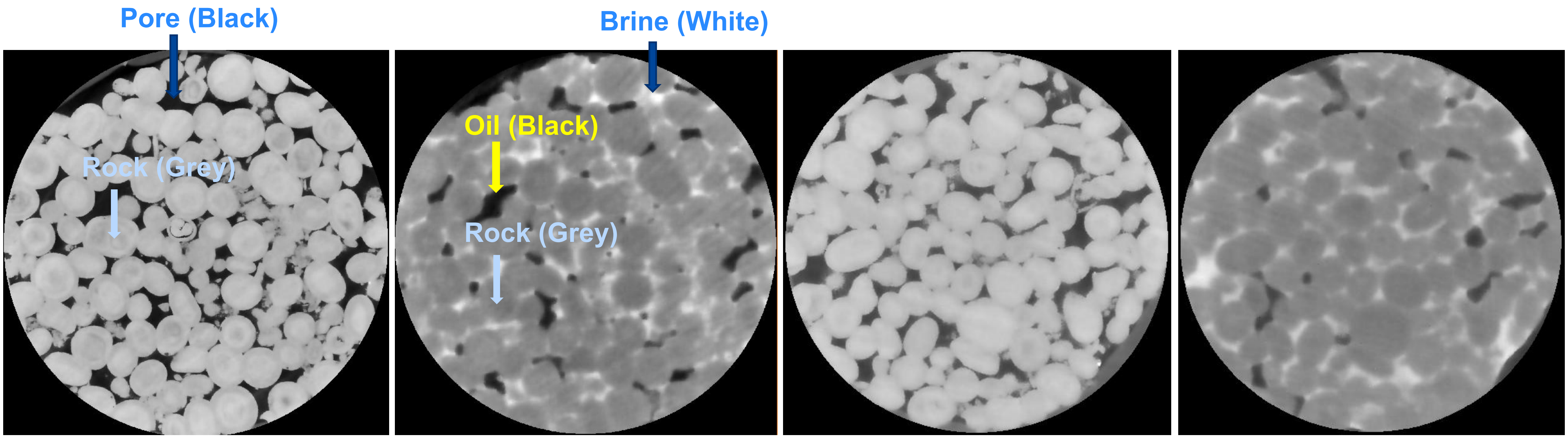


Water-Wet 2#



Oil-Wet

• Water-Wet Experiment



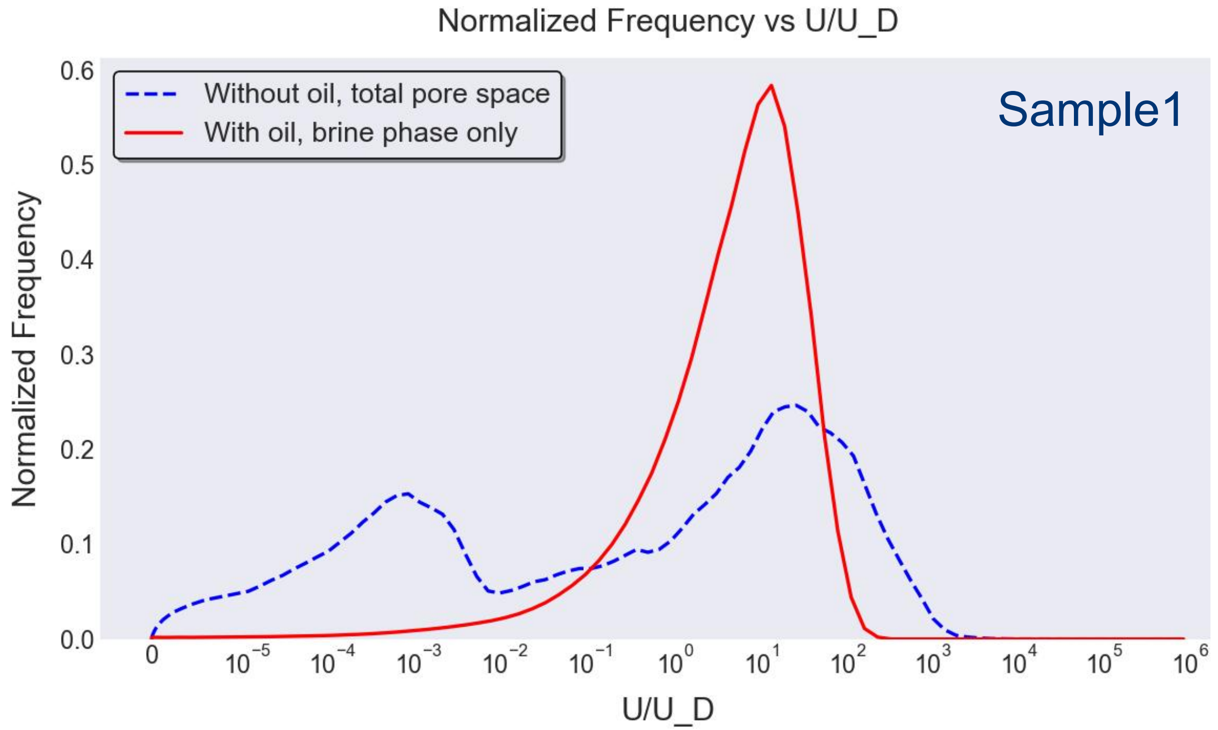
Sample-S1 | Sor= 40% | Sample-S2 | Sor= 28.1%

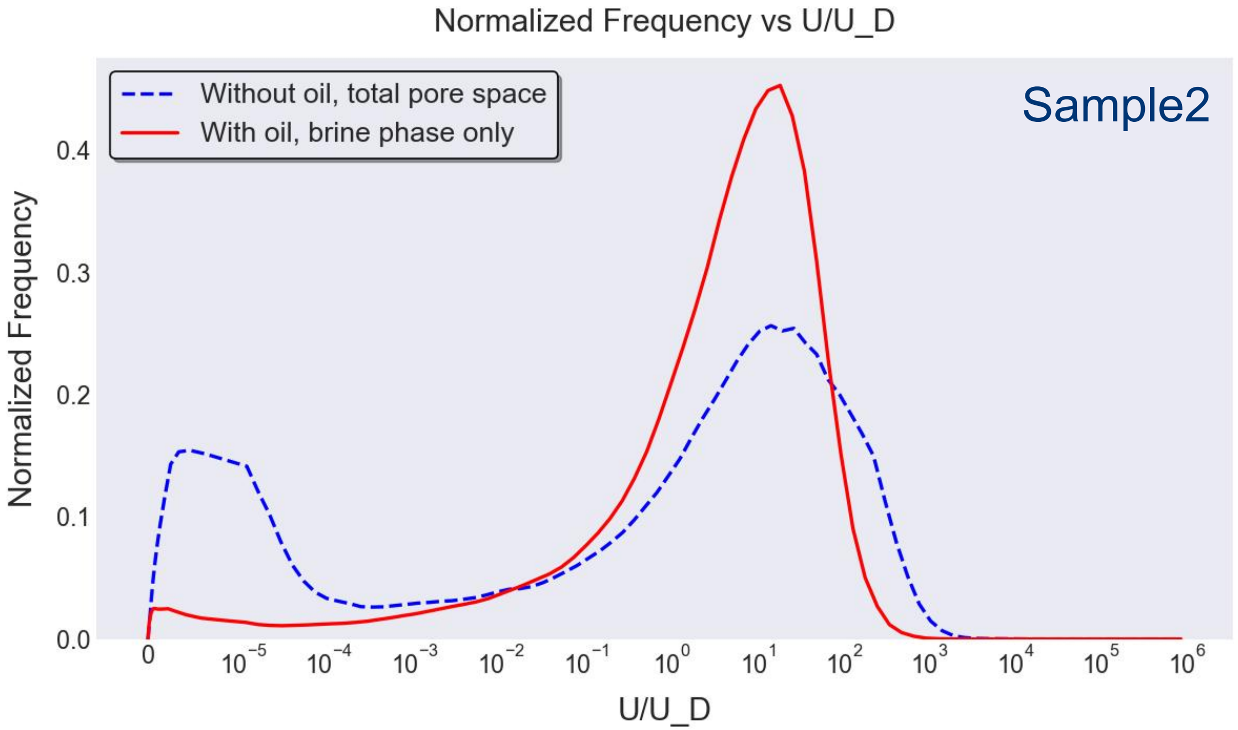
Rock Sample	S1	*S2*
Length, mm	120	120
Diameter, mm	60	60
Permeability(Darcy)	2.46±0.1	2.74±0.18
Micro-CT porosity (Marco-Porosity)	0.138	0.163
Oil Saturation	40%	28.1%

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

• Water-Wet Experiment

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

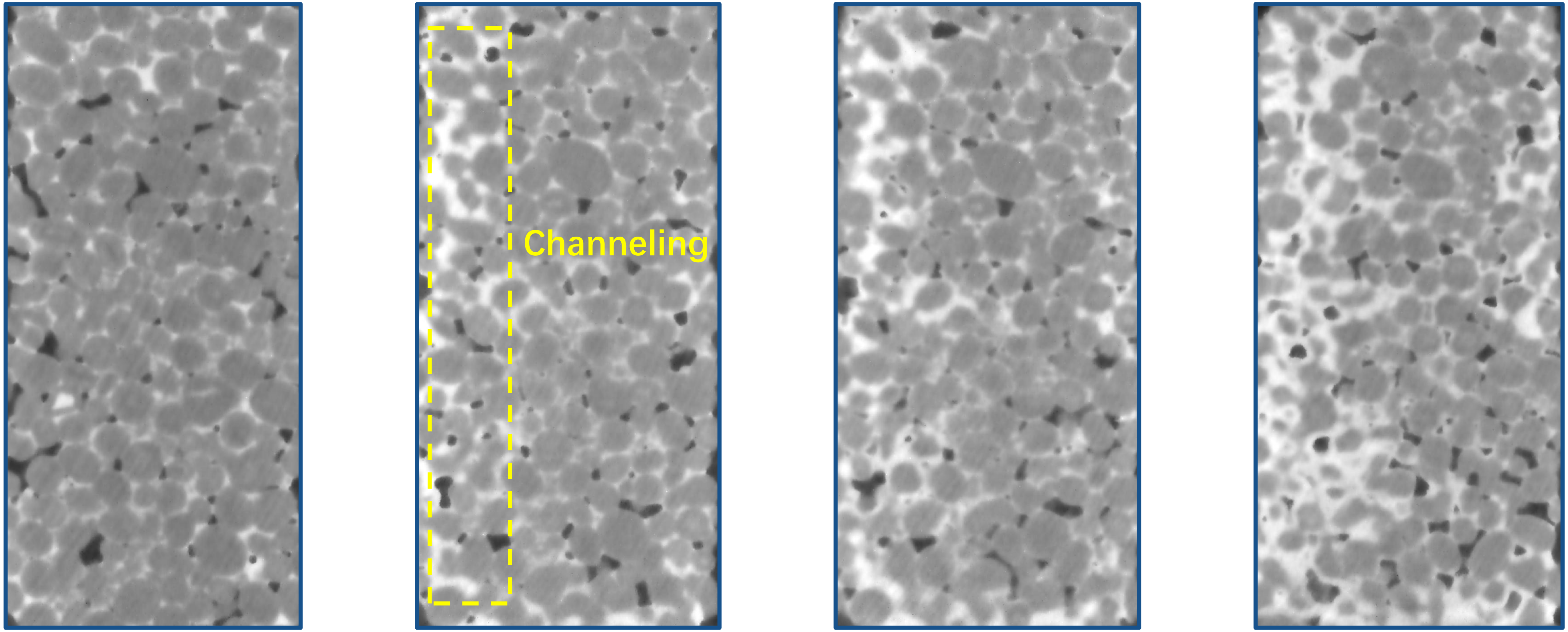




- The shape of the velocity distributions was used to characterize the physical heterogeneity.
- The introduction of oil phase makes the flow field heterogeneous (wider U distribution, smaller peak around normalized average velocity, more stagnant velocities).

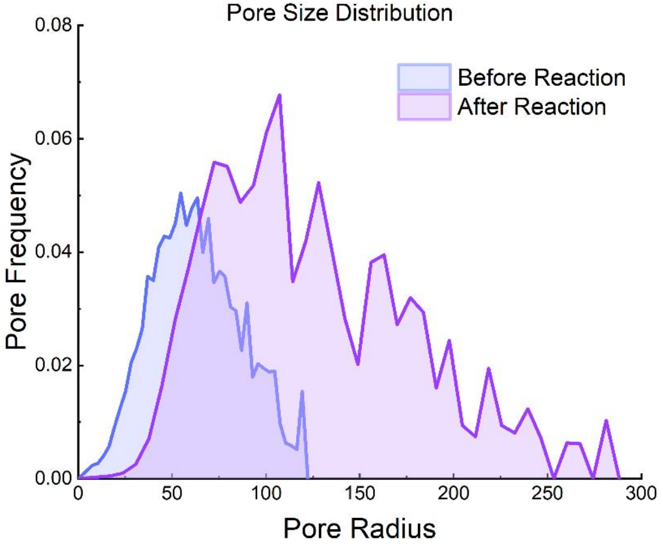
Water-Wet Experiment: Sample 1 (S_{OR}= 40%)

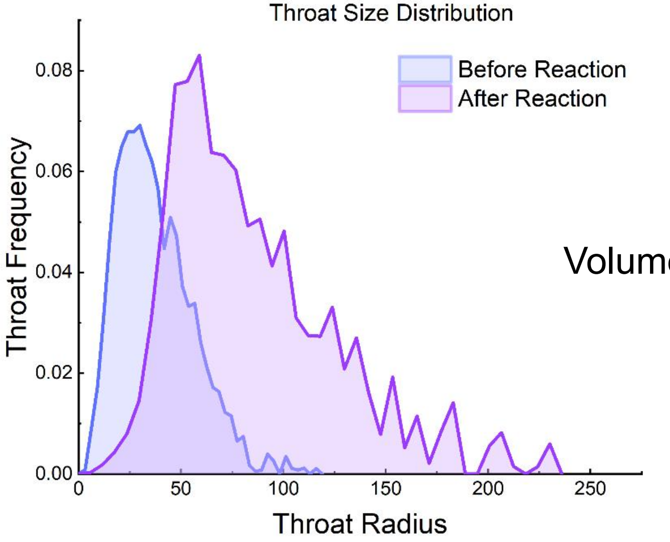
- Higher residual oil saturation facilitates the formation of dissolution channels.
- Reaction rates in regions of the core distant from the channel is significantly reduced.



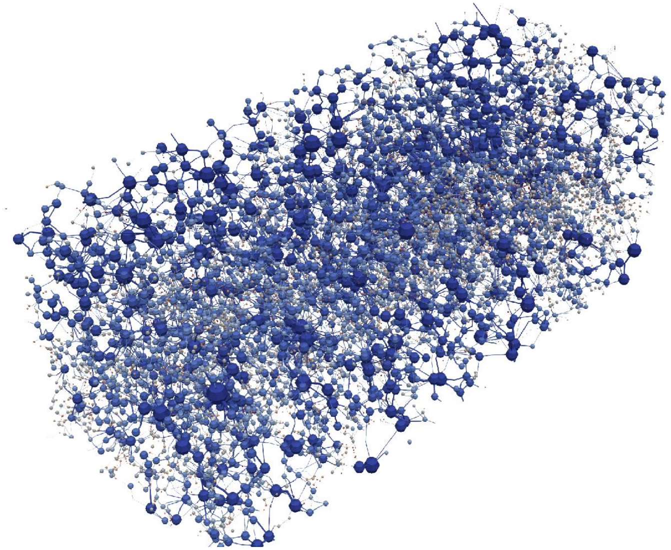
Before Reaction | 10 hours | 12 hours | 15 hours

Water-Wet Experiment: Sample 1 (S_{OR}= 40%)

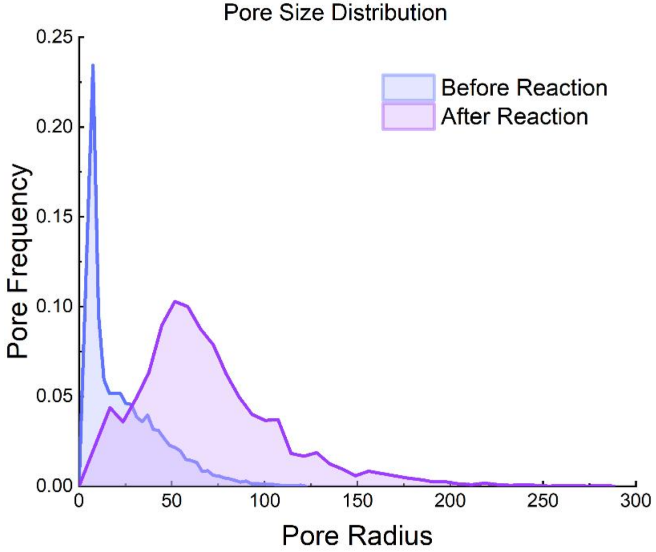


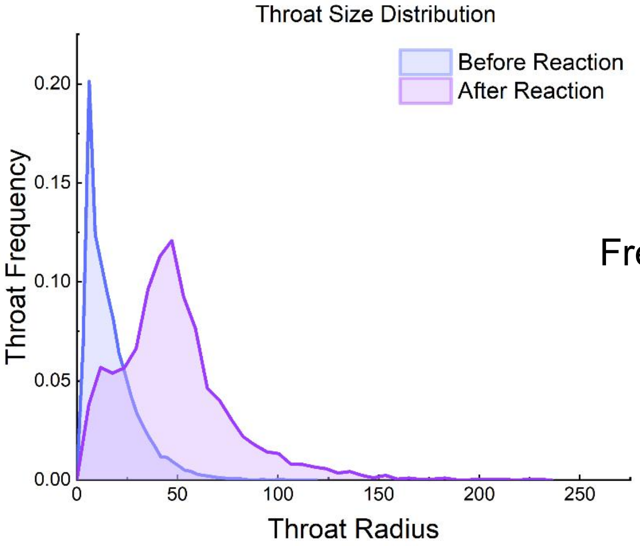


Volume Weight

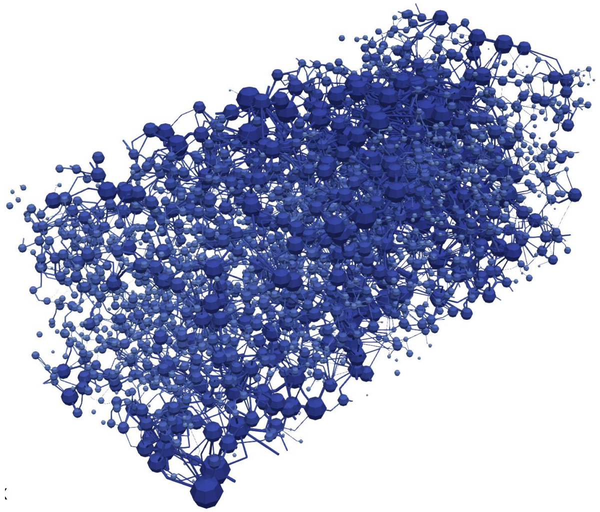


PNM Before Reaction





Frequency

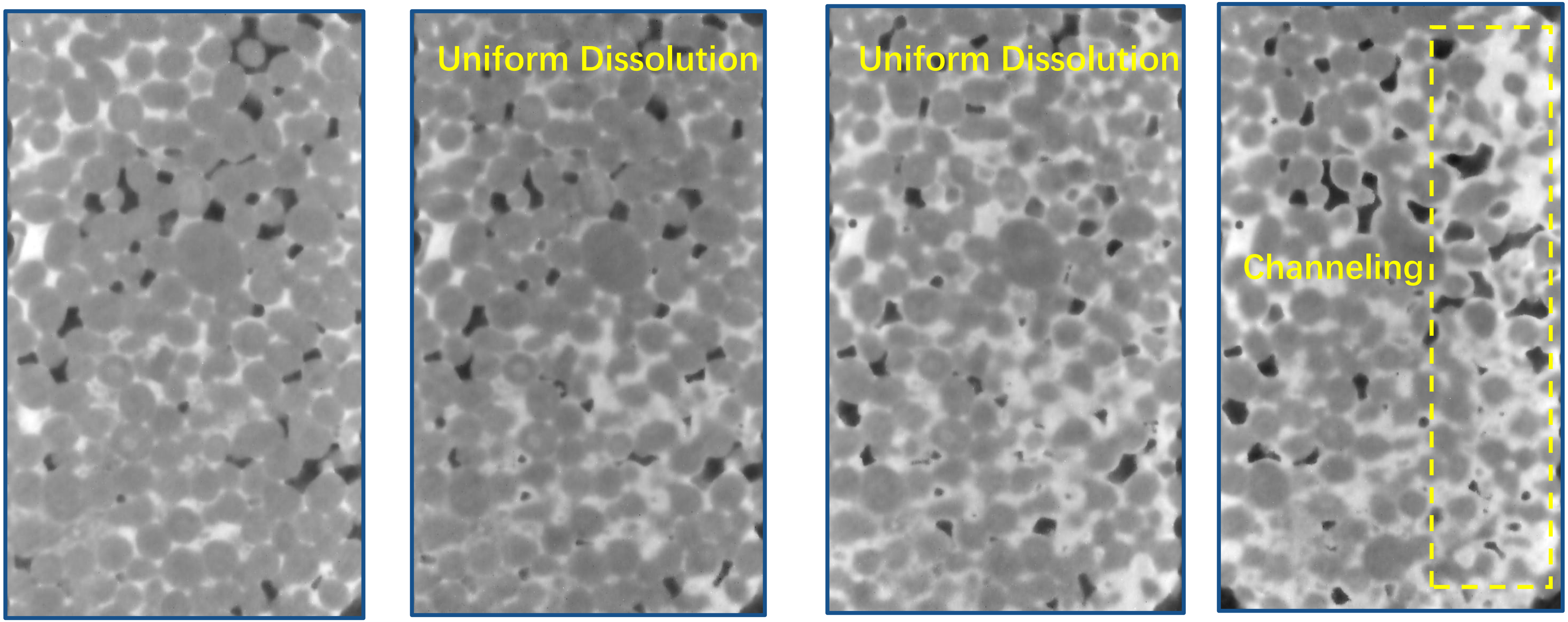


PNM After Reaction

- The pores and throats exhibit a significant shift toward larger sizes after the reaction.

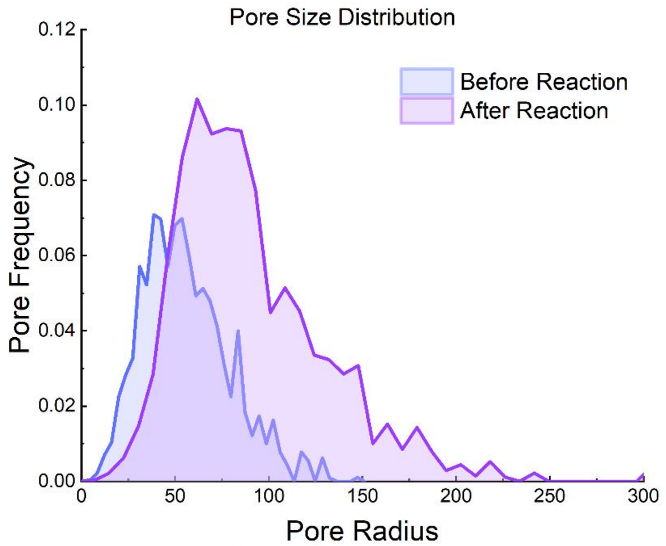
• Water-Wet Experiment: Sample 2 (S_{OR}= 28.1%)

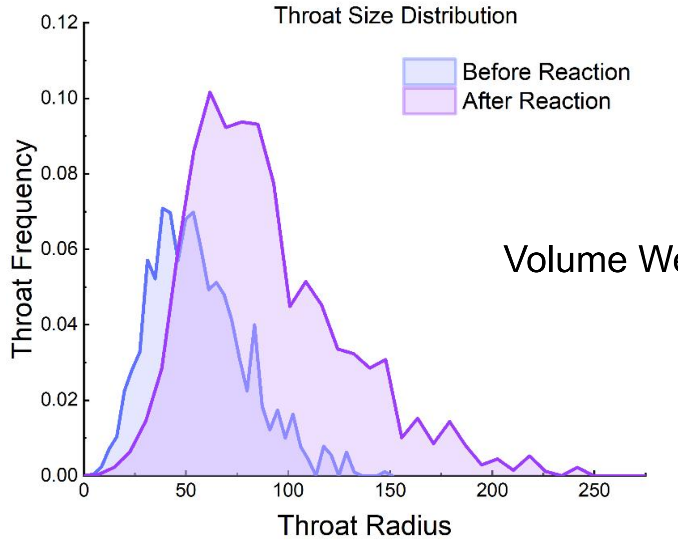
- Uniform dissolution during the early stages, with channels developing in the later stages of the reaction.



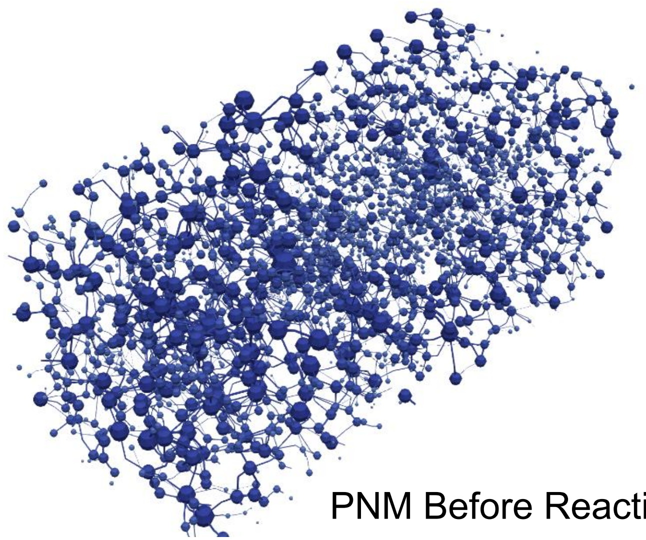
Before Reaction | 12 hours | 15 hours | 18 hours

• Water-Wet Experiment: SAMPLE 2 (S_{OR}= 28.1%)

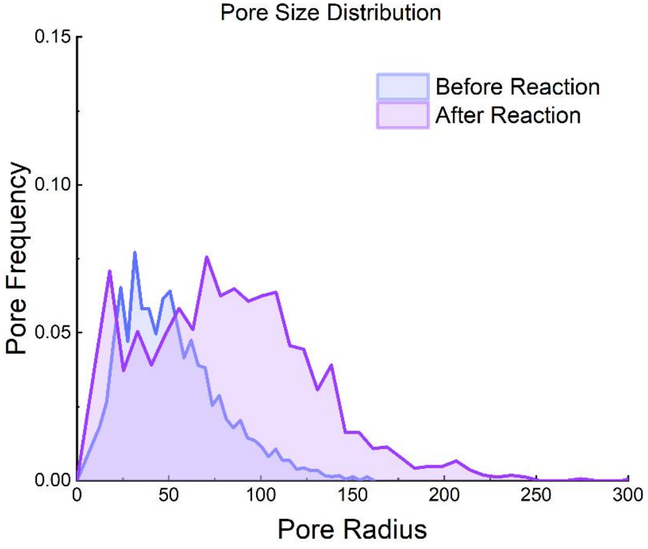


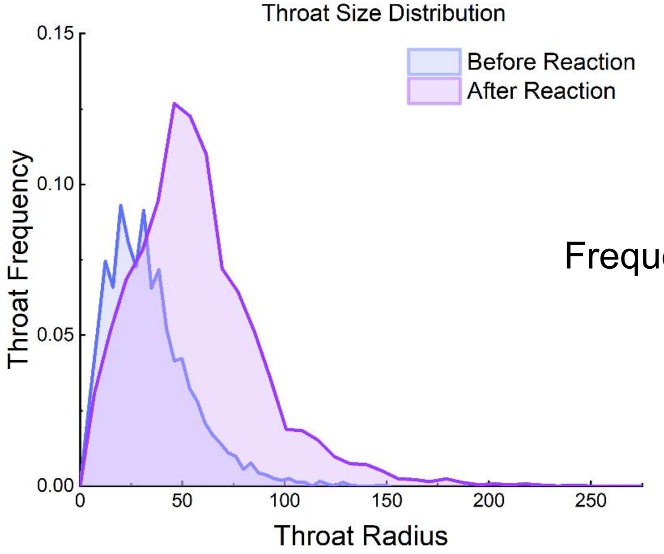


Volume Weight

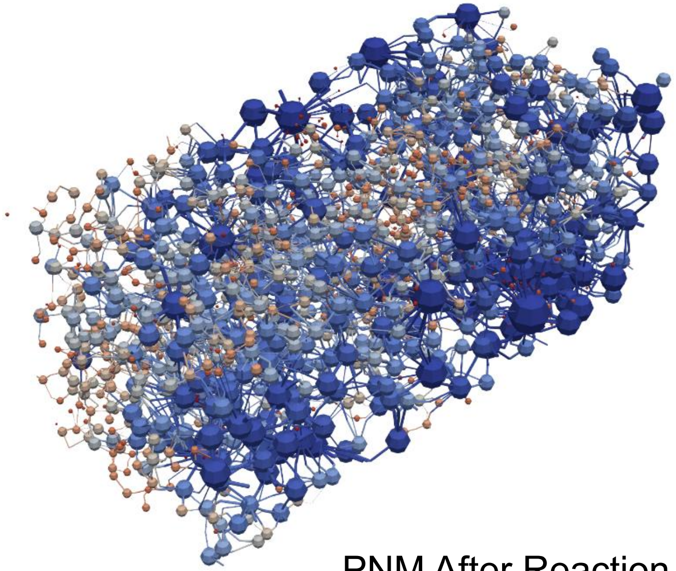


PNM Before Reaction





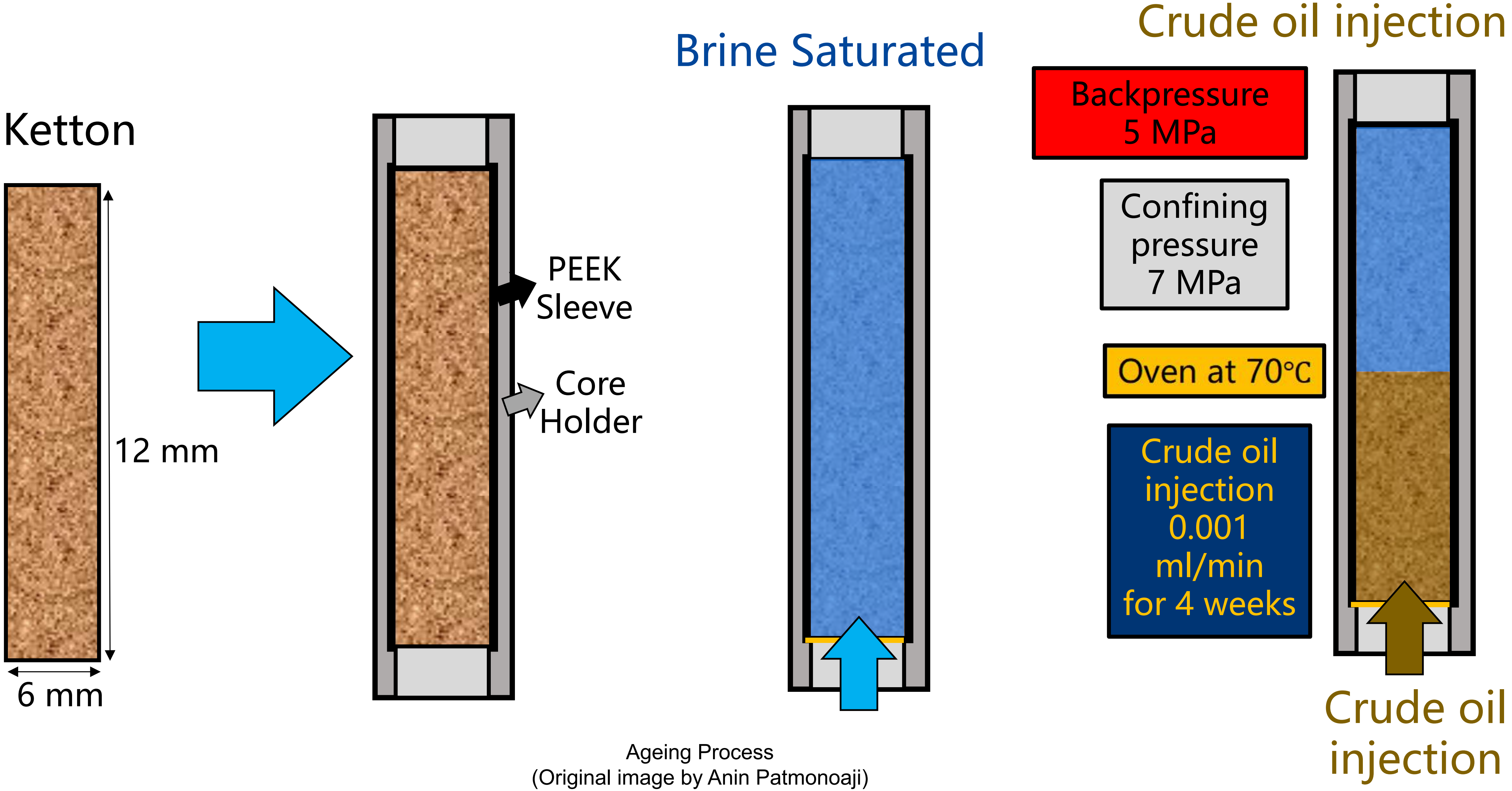
Frequency



PNM After Reaction

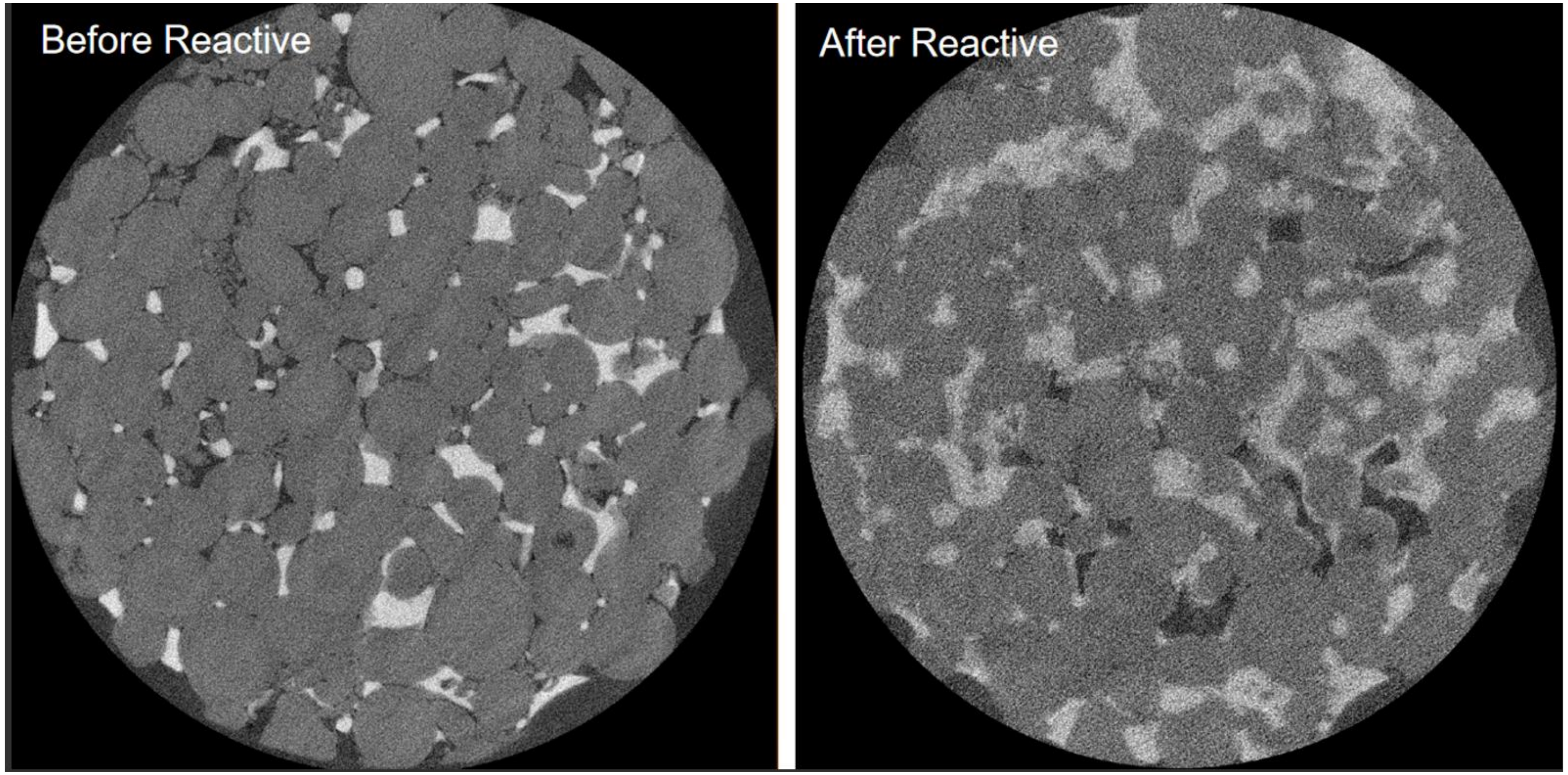
- The pores and throats shift to medium sizes, with limited expansion to larger sizes.

• Wetability Change: Crude Oil Ageing



Ageing Process
(Original image by Anin Patmonoaji)

Oil-Wet Experiment



- The reaction rate was noticeably slower compared to the water-wet samples.
- After dissolution, these samples appeared to become more water wet.

Summary

- The introduction of the oil phase increases the heterogeneity of the flow field.
- Higher residual oil saturation facilitates the formation of dissolution channels, while lower residual oil saturation tends to result in uniform dissolution during the early stages, with channels developing in the later stages of the reaction.
- Once dissolution channels are formed, reaction rates in regions of the core distant from these channels are significantly reduced.
- The reaction rate for oil-wet samples was noticeably slower compared to the water-wet samples.
- After dissolution, oil-wet samples appeared to become more water wet.

Future work

- Use direct numerical simulation (DNS) to quantitatively analyze transport heterogeneity based on the velocity distribution obtained from each scan.
- Quantitatively investigate the spatial distriution of oil during reactive transport processes.
- Quantification of reactive transport dynamics through effective reaction rate and Péclet & Damköhler Number.

[1] Wei, B., et al., ‘CO2 storage in depleted oil and gas reservoirs: A review’, Advances in Geo-Energy Research, 2023, vol. 9, pp. 76-93, doi: 10.46690/ager.2023.08.02.

[2] C. Noiriel and C. Soulaine, ‘Pore-Scale Imaging and Modelling of Reactive Flow in Evolving Porous Media: Tracking the Dynamics of the Fluid–Rock Interface’, Transp Porous Med, vol. 140, no. 1, pp. 181–213, Oct. 2021, doi: 10.1007/s11242-021-01613-2.

[3] R. Xu, R. Li, J. Ma, D. He, and P. Jiang, ‘Effect of Mineral Dissolution/Precipitation and CO2 Exsolution on CO2 transport in Geological Carbon Storage’, Acc. Chem. Res., vol. 50, no. 9, pp. 2056–2066, Sep. 2017, doi: 10.1021/acs.accounts.6b00651.

[4] Y. Al-Khulaifi, Q. Lin, M. J. Blunt, and B. Bijeljic, ‘Reaction Rates in Chemically Heterogeneous Rock: Coupled Impact of Structure and Flow Properties Studied by X-ray Microtomography’, Environ. Sci. Technol., vol. 51, no. 7, pp. 4108–4116, Apr. 2017, doi: 10.1021/acs.est.6b06224.

[5] X. Wang et al., ‘Multiscale wettability characterization under CO2 geological storage conditions: A review’, Renewable and Sustainable Energy Reviews, vol. 189, p. 113956, Jan. 2024, doi: 10.1016/j.rser.2023.113956.

[6] A. S. Abd and A. S. Abushaikha, ‘Reactive transport in porous media: a review of recent mathematical efforts in modeling geochemical reactions in petroleum subsurface reservoirs’, SN Appl. Sci., vol. 3, no. 4, p. 401, Mar. 2021, doi: 10.1007/s42452-021-04396-9.

[7] H. P. Menke, M. G. Andrew, M. J. Blunt, and B. Bijeljic, ‘Reservoir condition imaging of reactive transport in heterogeneous carbonates using fast synchrotron tomography — Effect of initial pore structure and flow conditions’, Chemical Geology, vol. 428, pp. 15–26, Jun. 2016, doi: 10.1016/j.chemgeo.2016.02.030.

[8] Y. Al-Khulaifi, Q. Lin, M. J. Blunt, and B. Bijeljic, ‘Reservoir-condition pore-scale imaging of dolomite reaction with supercritical CO2 acidified brine: Effect of pore-structure on reaction rate using velocity distribution analysis’, International Journal of Greenhouse Gas Control, vol. 68, pp. 99–111, Jan. 2018, doi: 10.1016/j.ijggc.2017.11.011.

[9] Y. Al-Khulaifi, Q. Lin, M. J. Blunt, and B. Bijeljic, ‘Pore-Scale Dissolution by CO 2 Saturated Brine in a Multimineral Carbonate at Reservoir Conditions: Impact of Physical and Chemical Heterogeneity’, Water Resour. Res., vol. 55, no. 4, pp. 3171–3193, Apr. 2019, doi: 10.1029/2018WR024137.

[10] H. P. Menke, B. Bijeljic, and M. J. Blunt, ‘Dynamic reservoir-condition microtomography of reactive transport in complex carbonates: Effect of initial pore structure and initial brine pH’, Geochimica et Cosmochimica Acta, vol. 204, pp. 267–285, May 2017, doi: 10.1016/j.gca.2017.01.053.

Acknowledgement

- Funding: We are grateful to the sponsors of the Resource Geophysics Academy, Imperial College London, for supporting this research.

- Supervisors: Dr. Branko Bijeljic, Prof. Martin Blunt

- Collaborators: Dr. Rukuan Chai, Dr. Anin Patmonoaji, Olatunbosun Adedipe, Anfal Al Zarafi

- Lab Staff: Dr. Edward Bailey, Dr. Vinchenzo Cunsolo