

# Trapping, Hysteresis and Ostwald Ripening in Hydrogen Storage

---

SEPIDEH GOODARZI

Supervisor: Prof. Martin Blunt  
Co-supervisor: Dr. Branko Bijeljic  
Department of Earth Science and Engineering  
January 2023

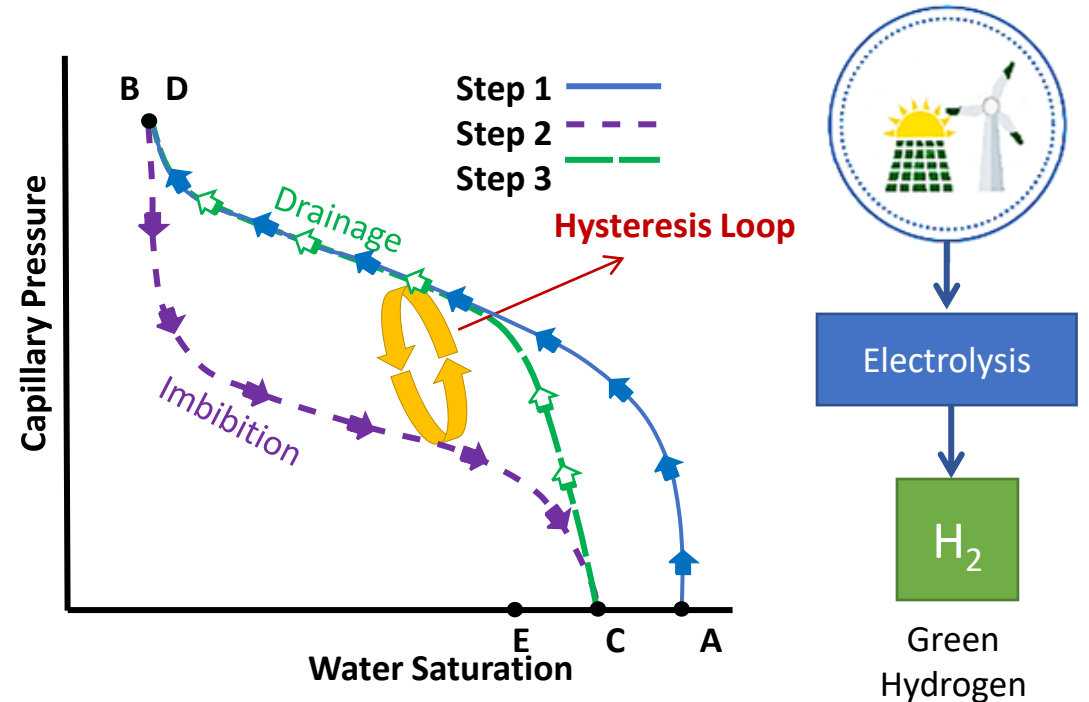
# Objectives of the project

---

- Fundamentals of dynamic hysteresis in multiphase flow
- A better characterization of multiphase flow properties, looking at both the effects of saturation path and flow rate.
- Potentially applicable to other situations including CO<sub>2</sub> and H<sub>2</sub> storage.
- Investigating hydrogen storage and use in different rock geometries.

# Background

- **Saturation, Interfacial area, Mean curvature and Gaussian curvature** give a full characterisation of geometry.
- Capillary pressure depends on both the saturation and saturation history.
- **X-ray imaging techniques** enable us to use this theory by measuring these four functions.
- **UHS** can be considered as a long-term energy storage solution (inject into the surface reservoir and withdrawn).
- **Porous formations** are good places for GT storage and use.



# Two-phase hydrogen experiment

- Investigate the hysteresis of a two-phase hydrogen-brine system at unsteady-state conditions.
- **Meso-scale, 12 mm diameter core and repeated H<sub>2</sub> and brine injection.**
- Lab-based micro-CT, using a Zeiss Versa XRM-500 X-ray microscope.

Table 1: Capillary number for unsteady state H<sub>2</sub>-Brine experiment,  $Ca_{ij} = \frac{\mu_i q_i}{\sigma_{ij}}$ .

Flooding Step	$Ca_{wg}$	$Ca_{gw}$	Flowrate(ml/min)
Water	5.8E-09	-	0.06
Gas	-	1.9E-07	2.00
Gas	-	3.9E-08	0.40
Gas	-	7.7E-09	0.08

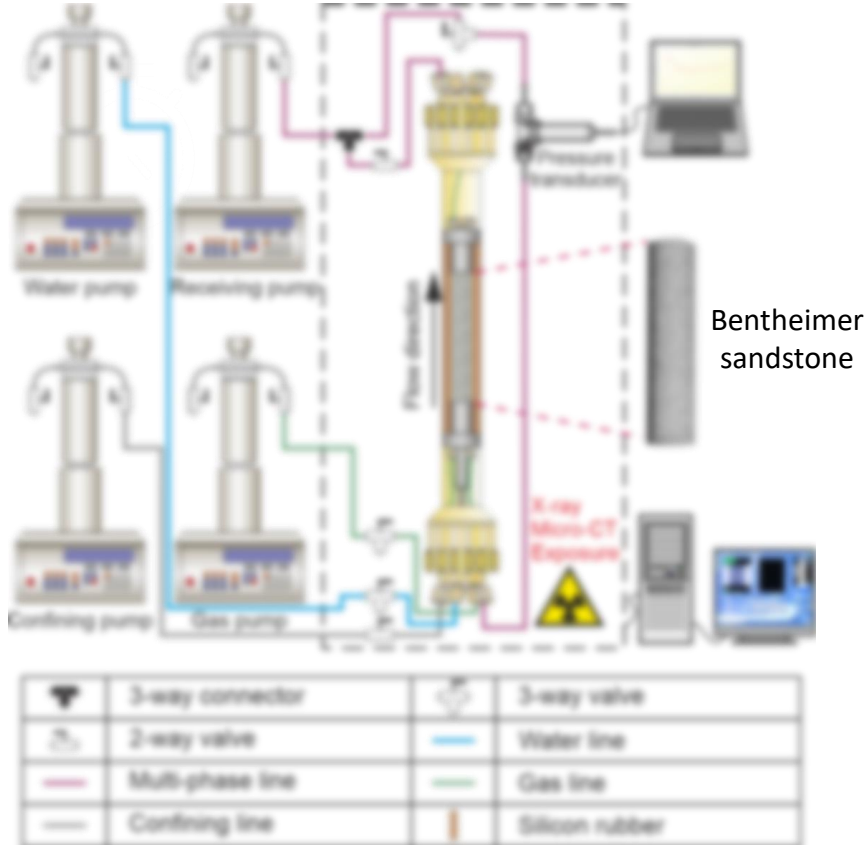


Figure 2: Schematic diagram for the two-phase gas-water experiment.

# Dry, normalised and segmented images

- Using Non-Local Means filter with Watershed segmentation technique.
- Image resolution is 5.86  $\mu\text{m}$ .

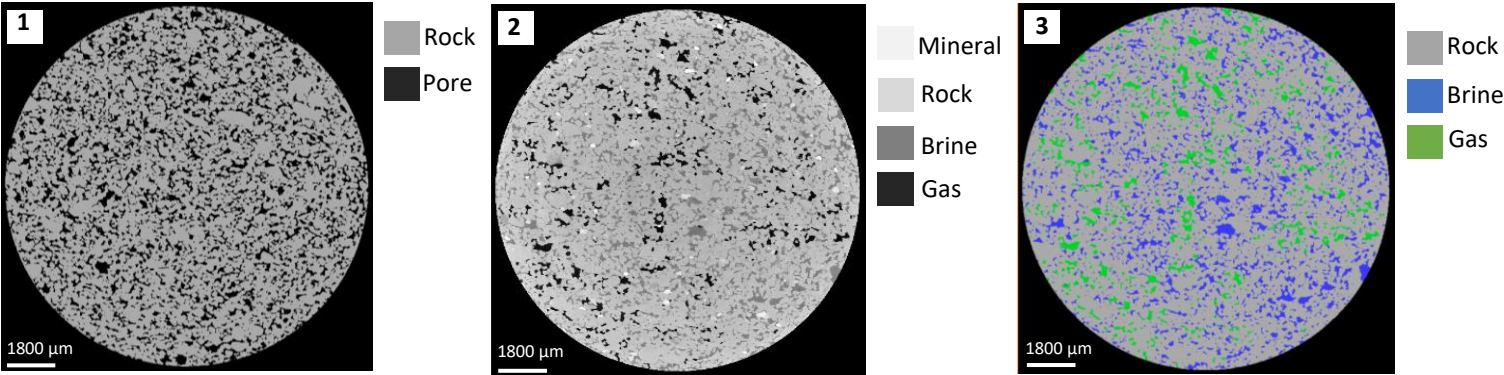


Figure 3: Dry(1), normalised(2) and segmented(3) two-dimensional slices of three-dimensional images of the Hydrogen-brine experiment.

Table 2: Procedure of the sample scanning.

Scan	Steps	Section
1 <sup>st</sup>	Dry scan	1-6
2 <sup>nd</sup>	Fully saturated	1-6
3 <sup>rd</sup>	<b>1<sup>st</sup> Drainage</b>	<b>1-6</b>
4 <sup>th</sup>	after 16hrs	2, 5
5 <sup>th</sup>	<b>1<sup>st</sup> Imbibition</b>	<b>1-6</b>
6 <sup>th</sup>	after 16hrs	2, 5
7 <sup>th</sup>	<b>2<sup>nd</sup> Drainage</b>	<b>1-6</b>
8 <sup>th</sup>	after 16hrs	2, 5
9 <sup>th</sup>	<b>2<sup>nd</sup> Imbibition</b>	<b>1-6</b>
10 <sup>th</sup>	after 16hrs	2, 5
11 <sup>th</sup>	<b>3<sup>rd</sup> Drainage</b>	<b>1-6</b>
12 <sup>th</sup>	after 16hrs	2, 5
13 <sup>th</sup>	<b>3<sup>rd</sup> Imbibition</b>	<b>1-6</b>
14 <sup>th</sup>	after 16hrs	2, 5
15 <sup>th</sup>	after 1 day	1-6

# Imaging of gas saturation for all cycles

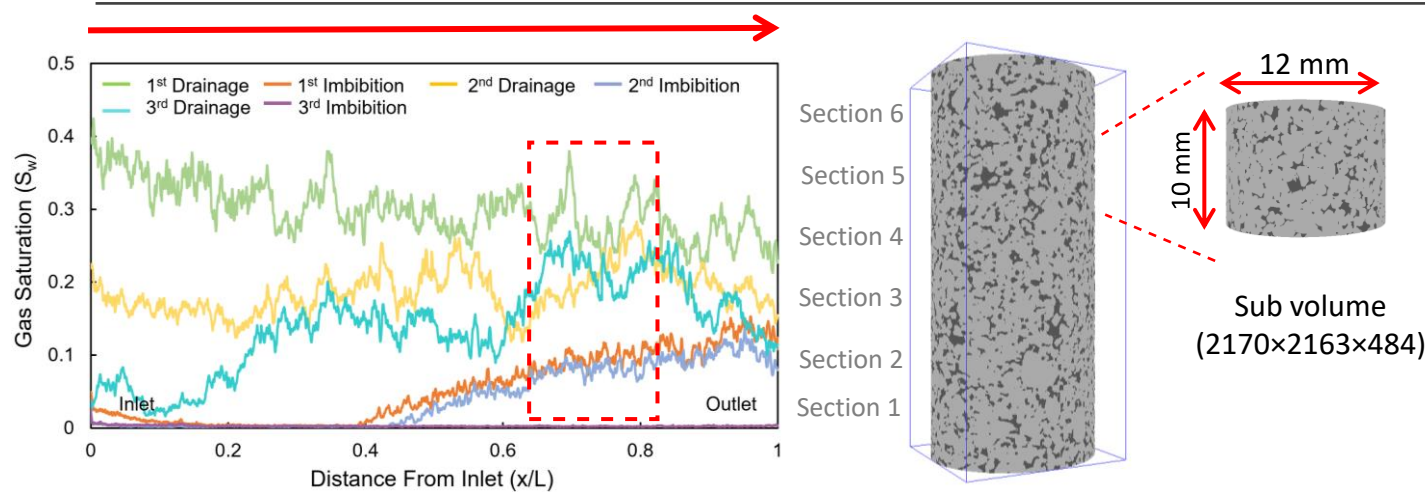


Figure 4: Fluid saturation profiles of gas during 3 cycles during imbibition and drainage.

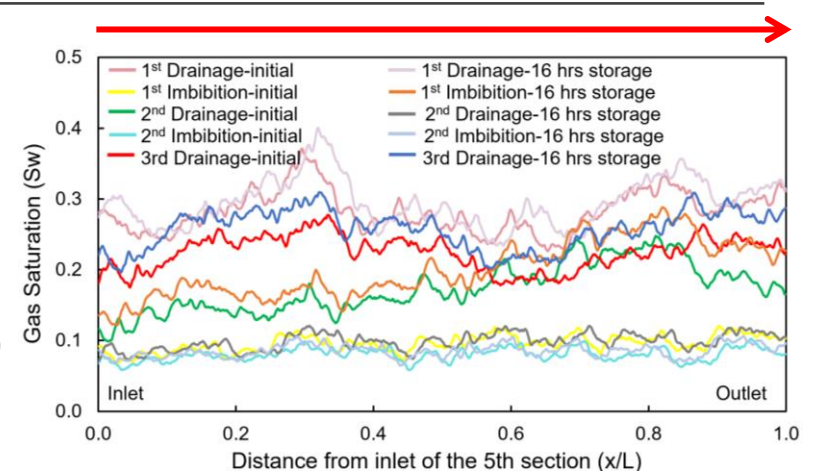
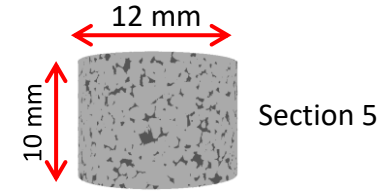


Figure 5: Saturation profiles of water during 3 cycles for a sub volume.

- Hydrogen is dissolved near the inlet when brine is injected.
- Gas saturation can increase after 16 hrs in the volume studied: gas may rise upwards from lower in the sample.
- Drainage (gas injection) leads to an increase in gas saturation.
- Not much change after water flooding.

# Analysis of discrete ganglia

## Ganglion size distribution (cycle 1)



- Rearrangement and tendency for some of the smaller bubbles to disappear and add to the larger bubbles but not necessary completely in a connected ganglion
- After imbibition one big ganglia begins to dominate the volume, which makes the withdrawal of hydrogen through a connected pathway possible.

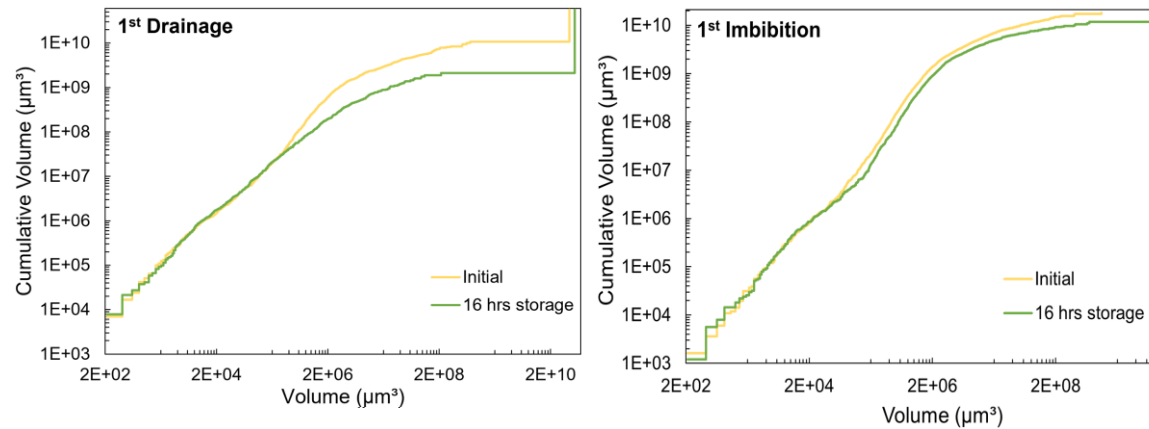
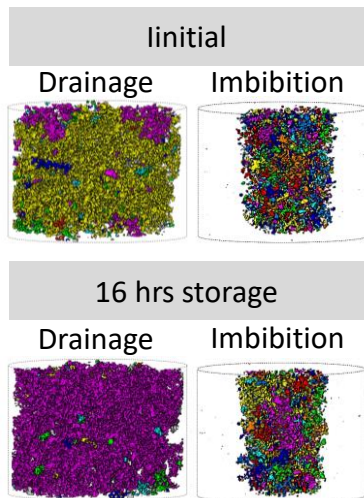
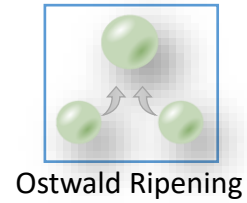
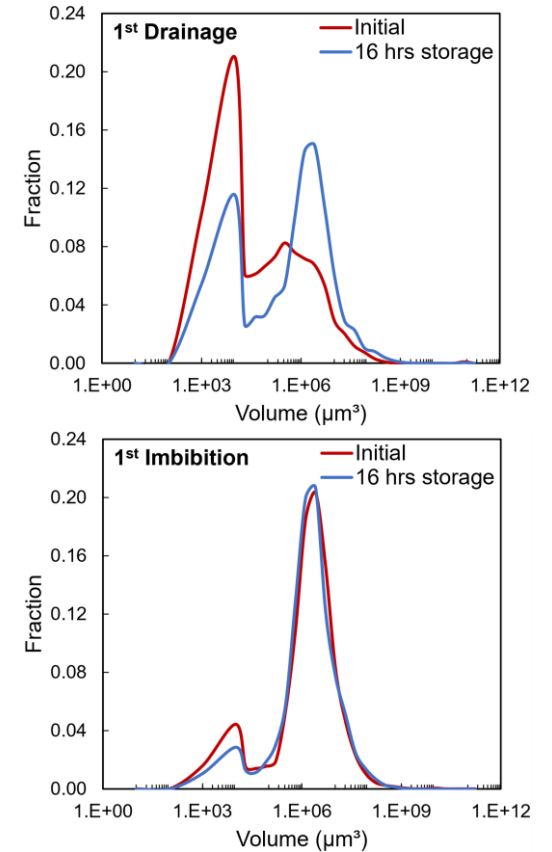


Figure 6: Ganglia size distribution of hydrogen-brine during first cycle for a sub volume.



# Analysis of discrete ganglia

Ganglion size distribution (cycle 2 & 3)

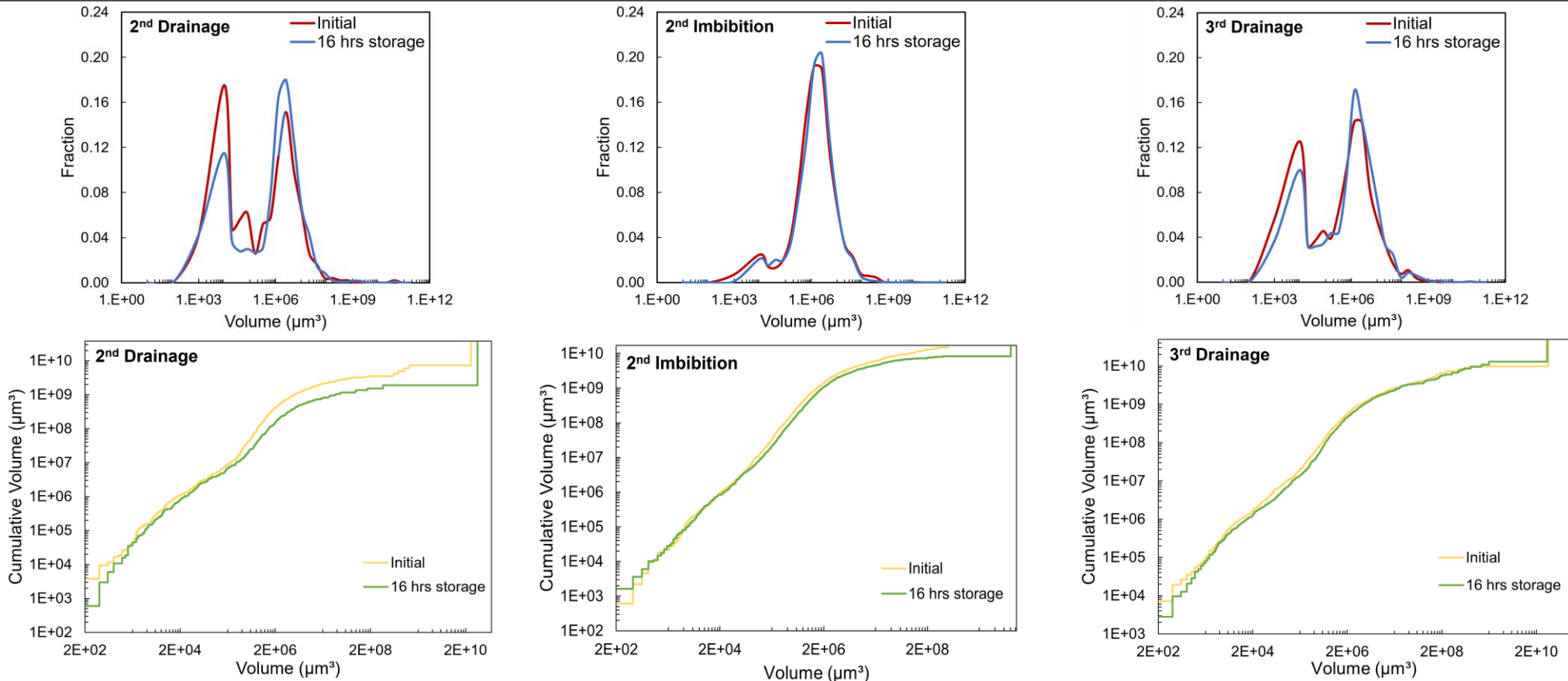
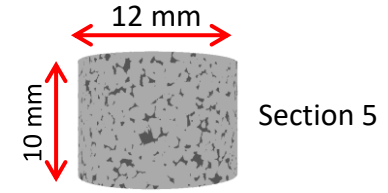
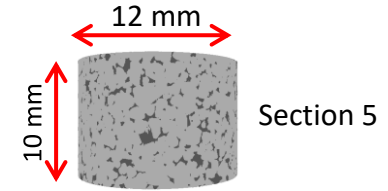


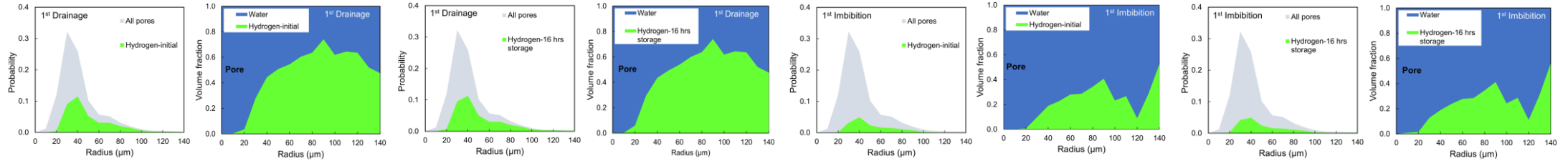
Figure 7: Ganglia size distribution of hydrogen-brine during second and third cycles for a sub volume.



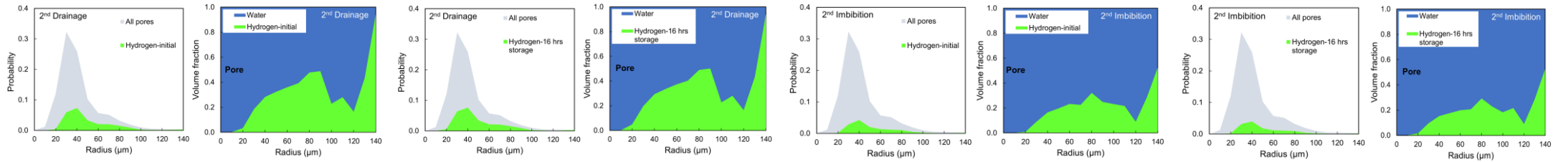
# Pore occupancy maps



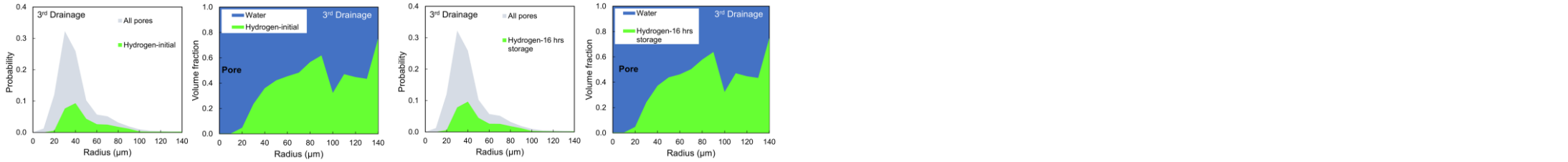
Cycle 1



Cycle 2

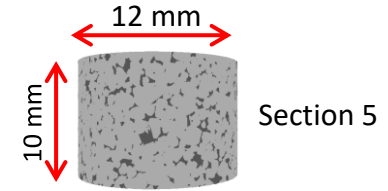


Cycle 3

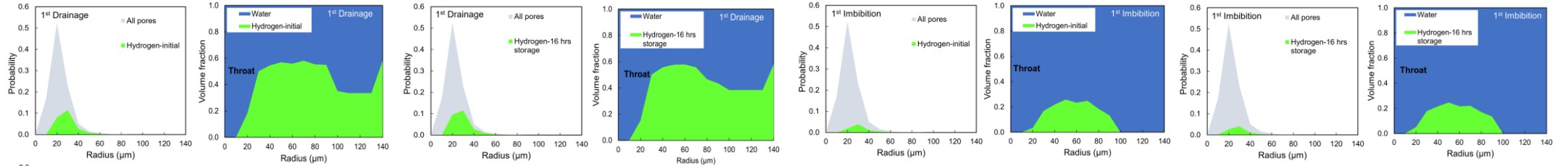


As expected, gas (shown in green) tends to reside in the larger pores. Both the actual distribution and occupied fractions are shown.

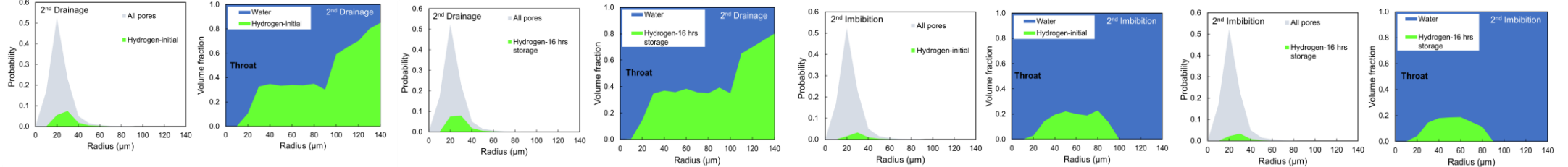
# Throat occupancy maps



Cycle 1



Cycle 2



Cycle 3

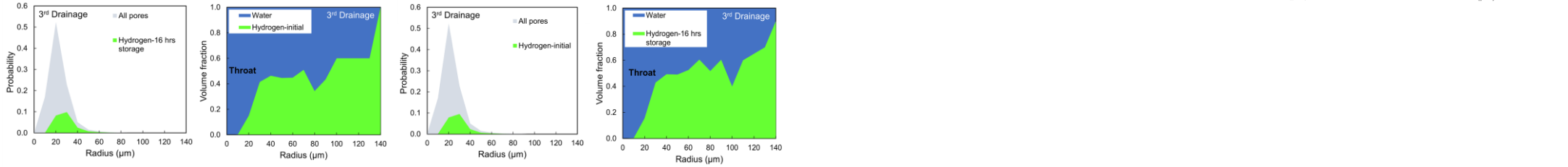
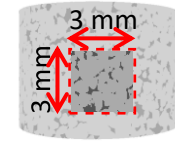


Figure 8: Histogram of the distribution of gas-filled pore elements at different stages of gas injection and the throat occupancy of the volume fraction at different radii.

# Measurement of interfacial area & capillary pressure analysis



Section 5  
Sub volume  
(500×500×500)

- Can't rely too much on pressure because of uncertainty in the measurement.
- Capillary pressure after drainage is slightly higher than after imbibition.
- No clear trend between initial and after waiting 16hrs, can go up or down.

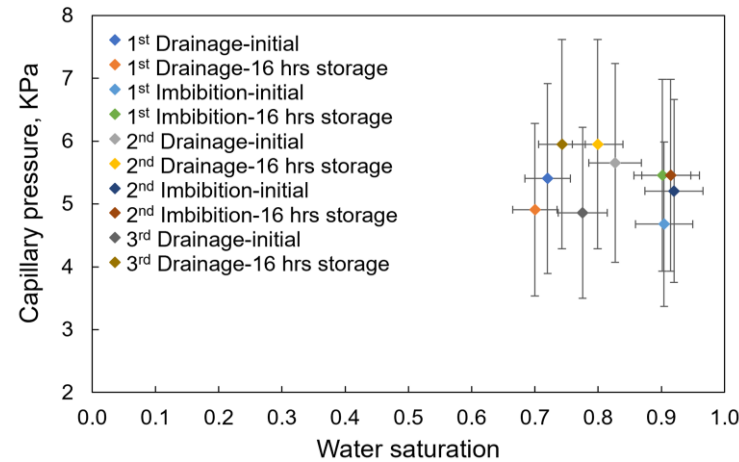


Fig 9: Capillary pressure measurements of each two phases for initial and after 16hrs.

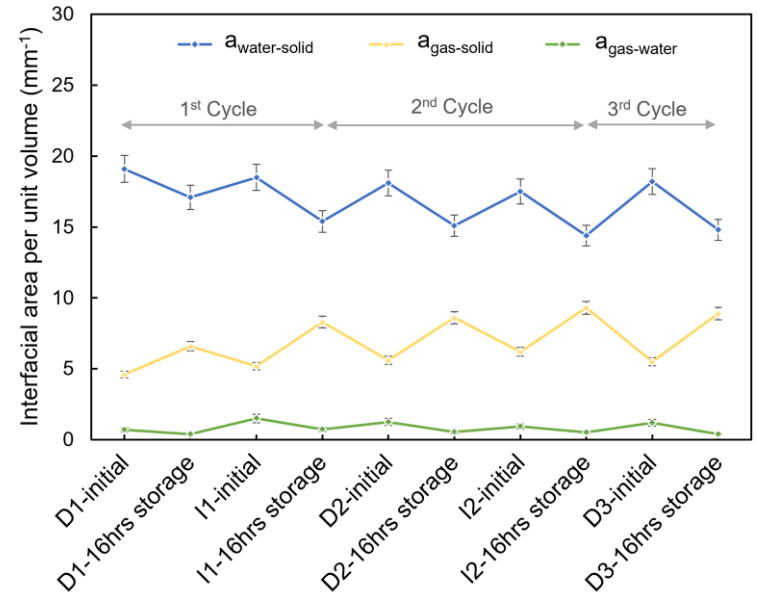


Figure 10: Interfacial area measurements of each two phases for initial and after 16hrs for 3 cycles

# Improvement of the experiment protocol

- ❖ Porous plate to allow a higher initial hydrogen saturation.
- ❖ Using imposed pressures to determine capillary pressure in drainage.
- ❖ Pre-equilibrated brine to reduce the effect of dissolution.
- ❖ Higher pressure, 10 MPa, to better represent reservoir conditions.

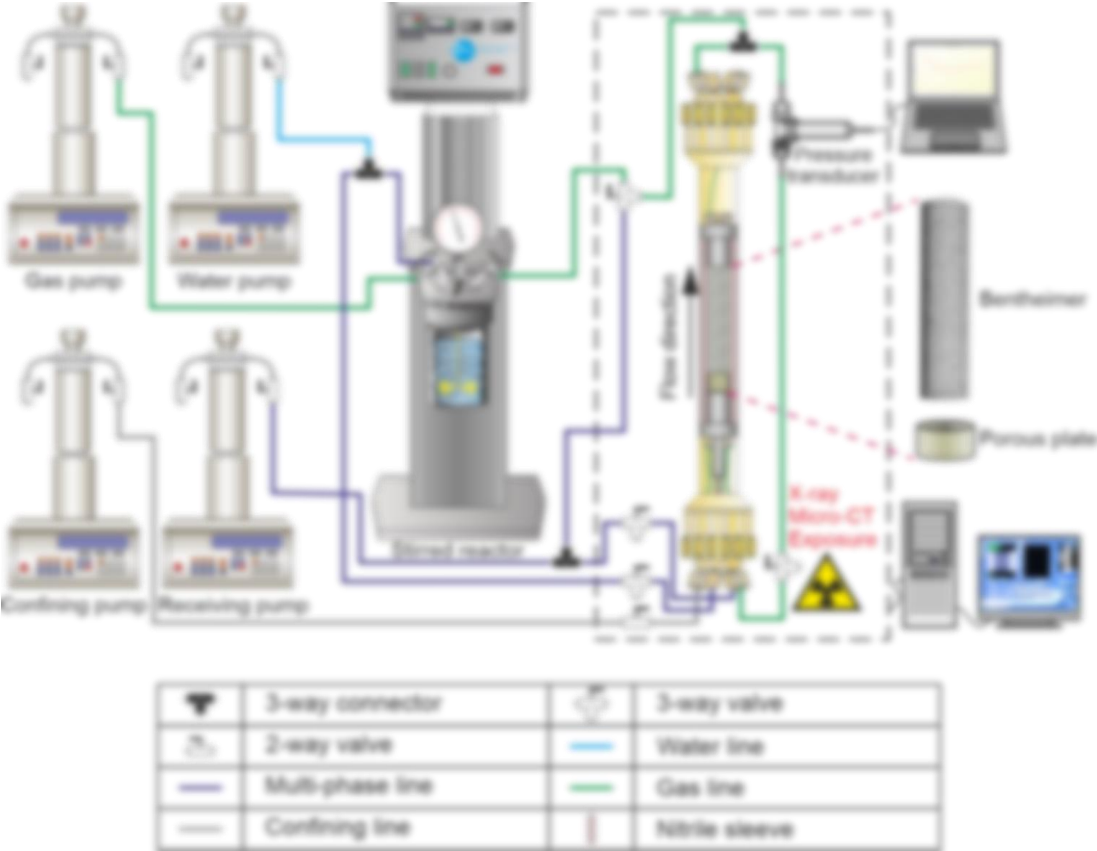


Figure 11: Schematic diagram for the two-phase gas-water experiment.

# Did the experiment work?

- High Initial gas saturation after hydrogen injection
- Uniform gas saturation after injection and withdrawal

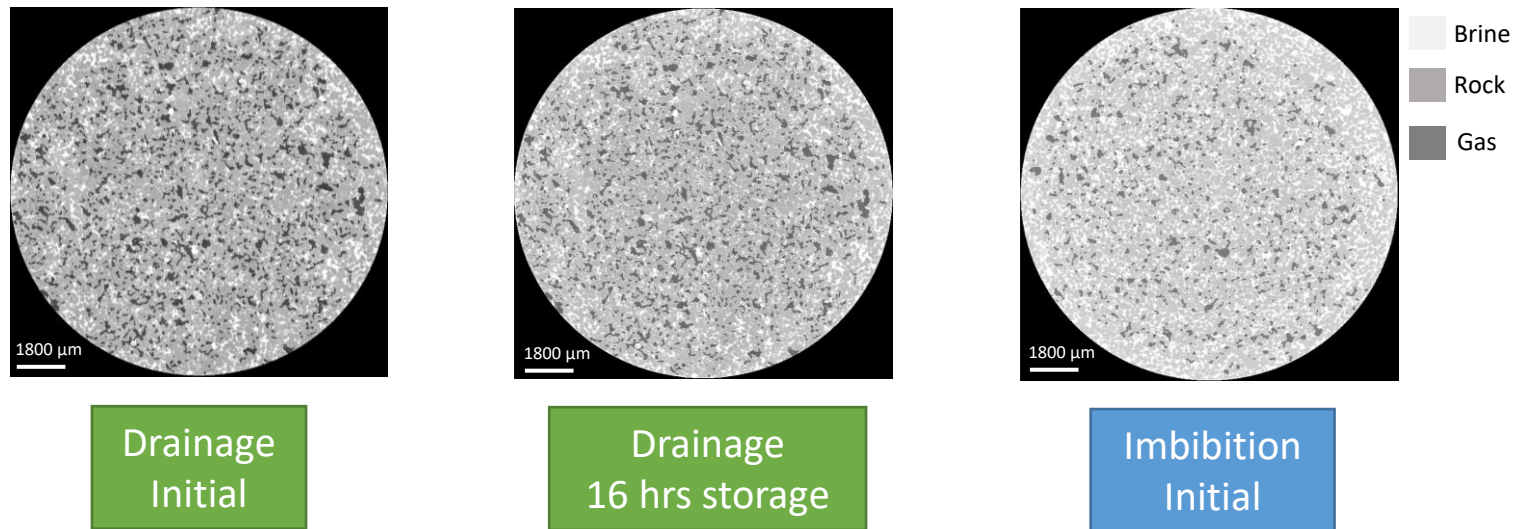


Figure 12: Dry two-dimensional slices of three-dimensional images of the Hydrogen-brine experiment.

# Conclusions

---

- Studying hysteresis during hydrogen injection is the main purpose of my work.
- The relationship between saturation, area, curvature and Gaussian curvature will be measured.
- Drainage and imbibition processes in 3 cycles for a hydrogen-brine system at the unsteady state condition have been investigated.
- The gas saturation was between 30% and 40% after each gas injection near the top of the sample
- Dissolving gas in brine changes the gas saturation along the sample.
- We saw the effect of Ostwald ripening: some of the smaller bubbles disappear and are rearranged to the larger bubbles but are not necessary in a connected ganglion.

# Future work

---

- I have looked at the carbonate sample for which I have performed a similar suite of experiments, and I am still analysing the results.
- Finishing the result analysis of Hydrogen-Brine experiment (porous plate).
- Preparing the procedure for a new set of experiment, different types of rock.
- How can we extend the experimental protocol to measure capillary pressure and relative permeability for a range of saturations, after allowing pore-scale equilibrium (Ostwald ripening)?

# Acknowledgements

---

- **Funding:**

Shell (Digital Rocks Phase2)

- **Supervisors:**

Prof. Martin J Blunt and Dr. Branko Bijeljic

- **Lab B109:**

Dr. Edward Bailey, Dr. Vincenzo Cunsolo, Dr. Yihuai Zhang and, Dr. Guanglei Zhang,  
Dr. Sajjad Foroughi





---

# Thank you