

UNDERGRADUATE RESEARCH PROPOSAL

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“Imbibition studies of capillary trapping and relative permeability of minerally heterogeneous sandstone samples”

1. Introduction

As the planet's climate data points to more alarming signals than ever before—higher global average temperatures and more extreme weather patterns—there becomes a necessity for robust technology and innovative forms of carbon sequestration. Injecting carbon dioxide into geologic porous media with high reactivity to supercritical CO₂ to form a mineral precipitate is a formidable option for capture, but logistical barriers and hyper specific in situ conditions complicate the process, necessitating the exploration of nano and microscale phenomena. The effectiveness of scCO₂ injection is largely dependent on capillary trapping and relative permeability, which serves as the concentration of the literature used to form this proposal.

2. Literature Review

“Co-sequestration of SO₂ with supercritical CO₂ in carbonates: An experimental study of capillary trapping, relative permeability, and capillary pressure”

Morteza Akbarabadi, Mohammad Piri

Using a Madison Limestone core sample, this paper examines the capillary pressure, capillary trapping, and relative permeability using steady-state drainage-imbibition. Implementing a scCO₂ + SO₂/brine/limestone system, they found that relative drainage permeability was low (0.04 H/m) where between 62.8% and 76% of the initial system was trapped in the capillaries of the rock. Additionally, unsteady-state primary drainage, imbibition and secondary drainage experiments were used to characterize the capillary pressure of the system. Compared to data collected in other experimental models, this system resulted in more residual trapping. An additional imbibition experiment was performed to measure residual scCO₂ rates with high brine flow rates, which resulted in a decrease of scCO₂ saturation.

“Water–rock interactions during a CO₂ injection field-test: Implications on host rock dissolution and alteration effects”

N. Assayag, J. Matter, M. Ader, D. Goldberg, P. Agrinier

Two push-pull test experiments were utilized to study the rates of in-situ CO₂-fluid-rock reactions at the Lamont-Doherty Earth Observatory. A non-reactive control test without CO₂ supplements the reactive test and was injected between chilled dolerite in the Palisades sill and underlying sediments of the Newark Basin. For the reactive test, quantities of dissolved inorganic carbon increase, shown by the dissolution of carbonate minerals. These tests confirmed that accurate DIC measurements can quantify the contribution of CO₂-fluid-rock reactions, which provides critical knowledge about capillary trapping potential in in-situ conditions.

“Drainage and imbibition CO₂/brine relative permeability curves at in situ conditions for sandstone formations in western Canada”

Stefan Bachu

Relative permeability characteristics is one of the single largest factors affecting CO₂ trapping rates, and predicting capillary trapping can be indicated by such. This paper examines three studies that examine relative permeability qualities of carbonate and sandstone aquifers at in-situ conditions in Canada. The results yielded a wide range of permeability data, running from less than 0.1 mD to 500 mD, suggesting that residual saturation is defined on rock characteristics (such as pore size) that are regularly measured in core analyses.

“Pore-scale study of capillary trapping mechanism during CO₂ injection in geological formations”

Uditha C. Bandara, Alexandre M. Tartakovsky, Bruce J. Palmer

In this study, a Smoothed Particle Hydrodynamics model was used to study capillary trapping and pore-scale displacement mechanisms of scCO₂. This approach was taken to investigate fluid displacement processes using a “first-principle” pore scale model. As a result of the experiment, the displacement patterns and capillary trapping is mainly dependent on capillary and gravity data. The trends in trapping demonstrated direct proportionality to the magnitude of gravity (larger gravity led to more trapping). Conversely, there was an inverse relationship between trapping and capillary numbers, as a high capillary number corresponds to relatively small trapping, due to buoyancy.

“A numerical study of dynamic capillary pressure effect for supercritical carbon dioxide-water flow in porous domain”

Diganta B. Das, Bhupinder S. Gill, Luqman K. Abidoye, Kamal J. Khudaida

This experiment examined numerical simulations of core-scale capillary pressure-saturation relationship for supercritical carbon dioxide. It considers the “dynamic capillary pressure effect” which finds that Pc-S relationships depend on both saturation and the time derivative with respect to saturation. This effect’s characterization in data is denoted by the dynamic coefficient (τ), and establishes the speed at which capillary equilibrium is met. The conditions of the simulation were in cylindrical porous domains, which had a 10 cm diameter and a 12 cm height. As a result, τ rose as saturation values decreased. However, these results were also affected by the temperature at 50% saturation. The conclusions suggest that time to reach capillary equilibrium is an integral factor of determining CO₂ capacity in an aquifer.

“Influence of capillary pressure boundary conditions and hysteresis on CO₂-water relative permeability”

Jiachao Ge, Xiaozhou Zhang, Jiachen Liu, Abdulmajeed Almutairi, Furqan Le-Hussain

This study performed drainage and imbibition injections into a Berea sandstone core and a sintered glass core in three injection stages (water, water-saturated CO₂, and CO₂ saturated water). Relative permeability data was collected by implementing the Johnson–Bossler–Naumann method, which was modified to factor capillary pressure boundary conditions. Its primary focus is history matching; the system is tampered with until the simulated data matches the experimental data. This adjusted method was utilized to increase the accuracy of initial guesses for history matching with respect to relative permeability. Experimentally, the CO₂-water relative permeability differed between the experimental and simulated values, which was a product of contact angle hysteresis.

“Capillary trapping of CO₂ in heterogeneous reservoirs during the injection period”

Naum I. Gershenzon, Robert W. Ritz Jr., David F. Dominic, Edward Mehnert, Roland T. Okwen

Specific to heterogeneous reservoirs, snap-off trapping and capillary pinning were used to study capillary trapping of CO₂. Snap-off trapping occurs during the post injection period, when imbibition of brine is prevalent at the tail of a buoyant CO₂ plume. This phenomenon is non-existent in heterogeneous reservoirs, as there is no imbibition during injection. However, this paper demonstrated that snap-off trapping was present in heterogeneous fluvial-type reservoirs during the injection period. The quantity of snap-off and capillary pinned CO₂ was shown to be dependent on the following characteristics: intrinsic permeability, irreducible water saturation, maximal residual CO₂ saturation, and capillary entry pressure barrier. Critically, the pinned amount is directly proportional to the low-capillary fraction of the rocks. Additionally, pinning data was larger in gravity-dominated flow than in viscous dominated flow.

“The impact of heterogeneity on the capillary trapping of CO₂ in the Captain Sandstone”

Catrin Harris, Samuel J. Jackson, Graham P. Benham, Samuel Krevor, Ann H. Muggeridge

This paper's analysis is focused on potential capillary trapping from natural rock heterogeneities in a study of cores from the Goldeneye formation of the Captain D Sandstone in the North Sea. This data is derived from imbibition processes that characterized the initial residual trapping relationships. The models show that the capillary heterogeneity ranged from 0-14% of the total trapped saturation. Results demonstrated a wide range of data; using the maximum experimental Land trapping parameter could increase trapping by a factor of 3, yet depending on the imbibition capillary pressure curve used, hysteresis could decrease heterogeneity trapping my maximum levels of 70%.

“A critical review of CO₂ mineral trapping in sedimentary reservoirs – from theory to application: Pertinent parameters, acceleration methods and evaluation workflow”

Sabber Khandoozi, Randy Hazlett, Milovan Fustic

This paper is a meta-analysis of carbon mineralization and capillary trapping in geologic porous media with the goal of isolating an effective workflow for the evaluation of such. In summary, the paper is concerned with the acceleration and practical implementation of mineralization by suggesting the following conditions for injection: increase of CO₂ solubility, careful selection of reservoirs and rocks with ultramafic tendencies, enhancement of reactive minerals surface area by fracturing. For the most effective in situ studies, pressure, temperature, caprock integrity, and comparative analysis are a necessity.

“Capillary trapping for geologic carbon dioxide storage – From pore scale physics to field scale implications”

Samuel Krevor, Martin J. Blunt, Sally M. Benson, Christopher H. Pentland, Catriona Reynolds, Ali Al-Menbali, Ben Niu

At reservoir conditions, capillary trapping was studied using X-ray microphotography. These images were produced to demonstrate pore-scale phenomena and create a database for flow modelling. In their findings, they confirmed that trapped saturations will be at minimum 10-30% of the pore volume of the rock. Current technological innovations focus on maximizing sweep and enhancing imbibition. The results of this experiment are consistent with the findings of other papers found in this literature review. There is still a relatively large amount of uncertainty concerning the effects of heterogeneity on capillary trapping in mixed-well systems.

“Micro CT and Experimental Study of Carbonate Precipitation from CO₂ and Produced Water Co-Injection into Sandstone”

Julie K. Pearce, Grant K. W. Dawson, Silvano Sommacal, Suzanne D. Golding

This paper examines the trapping mechanisms in a sandstone drill core from a reservoir proposed for CO₂ storage in reaction with scCO₂. Micro CT, QEMSCAN, and SEM were performed both before and after injection. The sandstone porosity was measured at 11.1% before the reaction and 11.4% after the reaction. The main proponent of the decrease in porosity was the precipitation of crystalline carbonate minerals in calcite and dolomite. Additionally, pore filling, bridging clays, and grains were observed after the injection of the solution.

“Capillary Pressure–Saturation Relations for Supercritical CO₂ and Brine in Limestone/Dolomite Sands: Implications for Geologic Carbon Sequestration in Carbonate Reservoirs”

Shibo Wang, Tetsu K. Tokunaga

This study focuses on capillarity and its hysteresis with relation to supercritical CO₂-water solutions. Capillary pressure (Pc)–saturation (Sw) relations of scCO₂ displacing brine and brine rewetting were used to understand CO₂ transport and trapping. Hysteretic drainage and imbibition curves were configured for a limestone sample at 45 degrees C under escalated pressures (8.5 and 12.0 MPa) in order to study supercriticality. Amounts of scCO₂ trapped increased with rising pressure, initial scCO₂ saturation, and time.

3. Research Proposal

With respect to the texts studied above, there are significant knowledge gaps and experimental challenges in the area of carbon mineralization in geologic porous media. Seeing as this lab focuses on in situ conditions, I only see it to be fitting that maintaining the characteristics of the sample of rock I will study remains of utmost importance, which is why I will be studying supercritical carbon dioxide. Similarly, I believe it is vital to examine heterogeneous samples of rock, as the majority of geologic compounds are not homogenous in nature and limiting mineralization to reservoirs allows for untapped potential for carbon dioxide sequestration. The characterization of heterogeneity in sandstones is a necessity outlined by the studies organized by Khandoozi and Krevor. Additionally, there is an extremely wide range of results in the data collected by Harris and Gershenzon. Thus, a thorough characterization of such when injected with scCO₂ is integral to increase our understanding of capillary trapping and relative permeability of the rock.

These gaps in study have led me to the following research questions:

How does mineral heterogeneity affect the quantity of residual CO₂ trapping in sandstones and various carbonates when the rate of injection is constant and when hysteresis is minimized?

If we are able to characterize the mineral heterogeneity of a sample, will such a characterization lead to a more thorough knowledge of mineralization in heterogeneous basins and reservoirs?

Answering these questions with thorough data analysis and experimentation will help to increase clarity of the impact of heterogeneity in capillary trapping and relative permeability, and leads me to a hypothesis whose goal is to isolate the impacts of heterogeneity on such. Thus, the hypothesis:

If testing two rock samples whose composition was the same for all but its mineralogy, a comparison of residual CO₂ in both rocks after injection of a CO₂ solution at the same conditions ought to be dependent on (a) the quantity of majority phase, (b) the quantity of minority phase, (c) the relative permeability of each, and (d) the characteristics of the rock in the minority phase. The goal of my experimentation is to draw conclusions that can isolate the effects of mineral heterogeneity in certain sandstone samples.

4. Proposed Tasks, Workflow, and Experimental Procedure

The goal of this study is to, first, get comfortable with imbibition technology, MATLAB, and the preparation of my samples. After initial data collection and organization, then I can proceed onto handling more delicate kinds of rock and develop ways to test the hypothesis and questions I have developed. More information is detailed in the workflow attached below.

PART I: Imbibition Overview

Imbibition includes the submersion of a cylindrical, porous rock sample in wetting fluid. Importantly, the sample must be previously “dry”, or saturated with a non-wetting fluid or gas such as air. A seal could be placed on any surface of the sample, as shown in [Figure 1](#), which would isolate relative permeability through one or a few faces of the sample. This technology can be used for testing capillary trapping, as it can study whether non-wetting fluids are stranded in pores after the insertion.

PART II: Experimental Procedure

This is a tentative experimental procedure, and is intended to be run for several trials. I plan to use this procedure to test the capillary trapping tendencies and to be adjusted as seen fit. Additionally,

this procedure is meant to be comparative; multiple trials will be run with rocks as identical as possible while holding a variable constant. Once data is synthesized from the first batch of experimentation, we can then adjust the variables in an attempt to isolate corollary parts of data. Generally, I plan to follow the procedure below:

1) Preparing the Sample

Using a core plug of a minerally heterogeneous sandstone sample, dry it and saturate it under controlled conditions. Dry the core at $\sim 60\text{--}80^\circ\text{C}$ under vacuum to remove all fluids (or use oven drying at 105°C depending on rock sensitivity). It is vital to prepare each sample under the same conditions, being careful to be as exact as possible. This ensures that the capillaries are as dry as possible for more effective trapping data.

2) Perform Drainage

Saturate the core with a 1-2 M brine under vacuum conditions and confirm saturation as close to 100% as possible using mass measurements. Then, inject the sample with air at a constant flow rate and continue until desirable initial saturation is reached. Allow the system to stabilize for a sizable amount of time (hours or days). We can adjust the concentration of the brine solution or even use water to imbibe the sample. It is likely that initial trials will simply use water in order to understand the trapping tendencies in a 'blank' state before using a brine solution to imbibe.

3) Perform Submersion

Allow the brine to be inserted back into the rock via the submersion of the sample into the solution. Measure how much non-wetting fluid is displaced and trapped. I plan for this test to be spontaneous rather than non-spontaneous, but like most aspects of this experimental procedure, we can adjust such as we begin to understand the tendencies of the sample.

4) Measure Capillary Trapping

Use MATLAB to monitor the volume of non-wetting fluid that comes out of the system and the weight change via a mass balance. The MATLAB codes can be pre-written and/or recycled from other experiments done in the lab. This will likely change on a trial by trial basis.

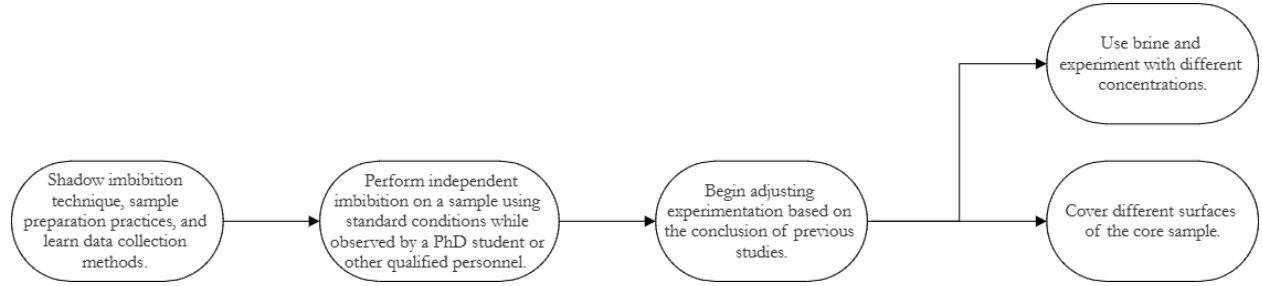
PART III: Micro CT Scanning

This aspect of the procedure is an intended supplement to the imbibition studies if the exploratory nature of this project warrants such. Most certainly, this will follow a substantial amount of imbibition experiments.

A CT scan can help to visualize the capillary trapping tendencies of a rock in a visual model, as shown in [Figure 2](#). Once we understand more about the trapping tendencies of a sample(s), we can

then insert fluid into the rock and depict the state of its capillaries and pores. To do this, I can utilize the scanner available at the Irving Medical Center and synthesize a file that would demonstrate the trapped air in the sample. This would likely require the help of those who are familiar with the technology, but I believe a physical model would be useful for the purposes of the study.

PART IV: Workflow Diagram



This workflow can be repeated several times and for different samples, picking up from step number two. Additionally, if I decide to utilize CT as a modelling option to experiment, I would attach it to the end of the workflow.

PART V: Data Analysis

This section functions as a list of possibilities for data configurations and graphical analysis of capillary trapping. The following can be used as seen fit for the experiment:

Calculations:

- ★ Washburn Equation - $L(t) = \sqrt{\frac{2\gamma\cos\theta}{\mu} \frac{t}{r}}$
- ★ Capillary Pressure - $p_c = \frac{2\gamma\cos\theta}{r}$
- ★ $\phi = (CT_{wc} - CT_{gc}) / (CT_w - CT_g)$
- ★ $S_w = (CT_c - CT_{gc}) / (CT_{wc} - CT_{gc})$

Graphs:

- ★ Saturation OR Mass vs. Time
- ★ Capillary Pressure vs. Saturation

The definition and units for each character are featured in the [appendix](#).

5. Conclusion

Conclusively, this proposal is certainly flexible, as its tentative workflow allows for a purely exploratory process. On the base level, I'm interested in using imbibition to explore mineral heterogeneity in sandstones and how such affects capillary trapping, but I will likely get a better sense of direction once I learn how to imbibe and start to get some initial data sets, which is to say: nothing is set in stone and it is likely we will make plenty of decision and adjustments as we go.

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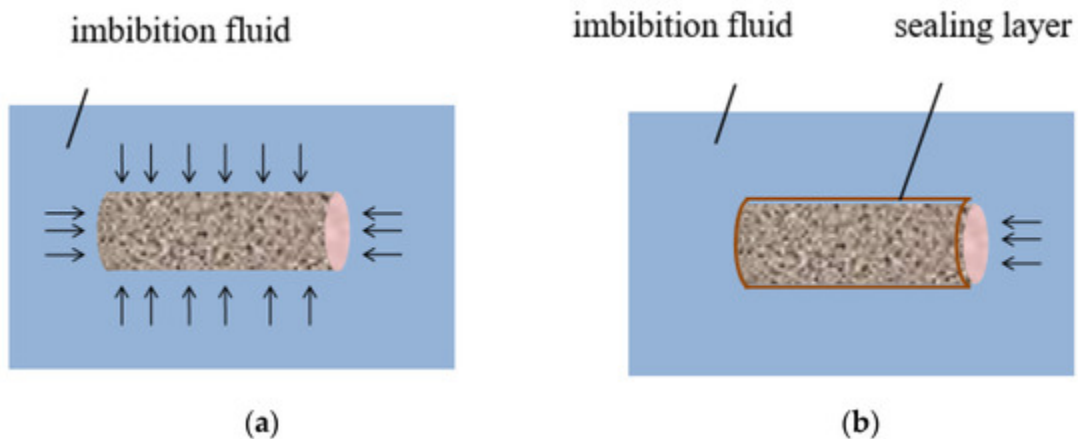
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Appendix

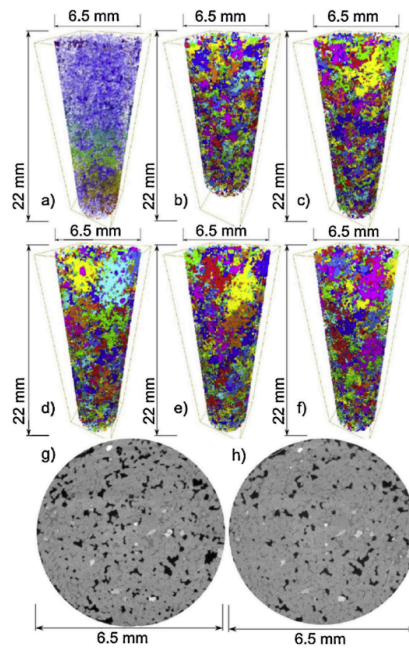
Figure 1:



The figure models the basic principles of the submersion portion of imbibition in Feng's paper. To be noted, sealing layers can be placed on any of the surfaces of the core sample.

Figure 2:

This figure is sourced from the paper of Krevor et. al. and demonstrates a sample CT scan of a rock after it has been injected by brine. It serves as a visualization for capillary trapping tendencies in a sample.



Data Analysis Characters:

L(t): penetration distance (m)

γ : surface tension (N/m)

θ : contact angle

μ : viscosity (kg/ms)

r: pore radius (nm)

Φ : porosity (%)

S_w : saturation (%)