

Imperial College London

Micro-CT imaging of multiphase flow at steady state

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Abstract

Pore-scale imaging provides an effective way to understand multiphase flow in porous media at the pore scale and provides benchmark data for larger-scale modelling. In this thesis, an X-ray tomography-based experimental and image analysis method was devised for investigating the relative permeability and associated pore occupancy during steady-state two-phase flow in a sandstone and a complex carbonate, and to identify dynamic flow regimes from low to high capillary numbers.

First, a two-phase flow experiment at different fractional flows at steady state was conducted on a Bentheimer sandstone sample at mm-scale using a laboratory micro-CT scanner at both low and high capillary numbers. An intermittent flow, defined as the regions occupied by oil and water alternately, was observed at high capillary number during the hour-long scan time. Differential imaging method was used to quantify the fraction of the intermittency in pore space. Imbibition relative permeabilities and fractional flow curves were obtained with precise differential pressure measurements which compared well with literature measurements on larger cm-scale cores.

This method was then expanded and applied into a micro-porous Estailades carbonate using X-ray micro-tomography. Differential imaging method was applied to (i) distinguish micro-pores and quantify micro-porosity; (ii) determine fluid pore occupancy in both micro-porous regions; and (iii) identify the intermittency in macro-pore space. The brine saturation and relative permeabilities were obtained, which were impacted by the presence of water-wet micro-porosity which provides additional connectivity to the phases. Pore and throat occupancy of oil, brine and intermittency were obtained from images at different fractional flows using the generalized pore network extracted from the image of macro-pores. The intermittent flow was observed at high capillary number and was predominantly located in the small and intermediate size pores and throats.

As laboratory-based instruments take around 1 hour to acquire an image, meaning that intermittency can only be observed indirectly, fast synchrotron X-ray micro-CT was used to capture changes in occupancy over time scales of approximate 1 minute. Thus, fluid behaviours as a function of flow rate (capillary number) at a fixed fractional flow of 0.5

were studied during steady state multiphase flow through a Bentheimer sandstone using synchrotron X-ray micro-tomography. Combining the fluid rearrangements observed from the continual scans taken every 60s with the pressure differential provided by the sensitive differential measurements, three flow regimes were classified: capillary-dominated flow, onset of dynamics, and intermittent flow. The pressure differential has a linear relationship with flow rate during the first two flow regimes. However, there is a power-law scaling of flow rate with pressure drop across the system during the intermittent flow regime. We observed that when there is sufficient energy to allow the oil to short circuit some pores and throats at high capillary number, the overall flow resistance decreases, meaning that oil moves along pathways that intermittently disconnect and reconnect.

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24th June, 2019

Declaration

I declare that this thesis, *Micro-CT imaging of multiphase flow at steady state*, is entirely my own work under the supervision of Prof. Martin J. Blunt and Dr. Branko Bijeljic. The work was performed in the Department of Earth Science and Engineering at Imperial College London. All published and unpublished material used in the thesis has been given full acknowledgment. This work has not been previously submitted, in whole or in part, to any other academic institution for a degree, diploma, or any other qualification.

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List of Publications

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Nomenclature

Symbol	Description
A	Cross-section area of flow domain [m ²]
Ca	Capillary number
Eu	Euler number
f_w	Wetting phase fractional flow, $f_{nw} + f_w = 1$
g	Acceleration due to gravity [m/s ²]
I	X-ray attenuated intensity
I_0	Original X-ray attenuated intensity
$i = w, nw$	Wetting phase (brine, de-ionised water), non-wetting phase (n-decane)
k, k_{abs}	Absolute permeability [m ²]
$k_{r,i}$	Relative permeability to w and nw phase
L	Length of sample, parallel to flow [m]
$n(s)$	The number of clusters of volume s
ΔP	Viscous pressure drop [Pa]
P_c	Capillary pressure [Pa]
Q_i	Flow rate [mL/min]
q_i	Darcy velocity [m/s]
r	Pore throat radius [m]
S_i	Saturation of phase i
t	Time
x_i	The thickness of solid object i during scan
$\beta_0, \beta_1, \beta_2$	Betti numbers
θ	Contact angle
μ_i	Dynamic viscosity [Pa·s]
μ	X-ray linear attenuation coefficient
ρ_i	Density [kg/m ³]
σ	Interfacial tension between w and nw phase [N/m]
τ	Fisher exponent
ϕ	Porosity

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Chapter 1

Introduction

Multiphase flows are encountered in many industrial and geophysical applications, such as carbon dioxide sequestration and oil recovery, groundwater management, blood flow in capillary vessels, catalyst supports used in automotive industry, bubble generation in microfluidics and many more. In this study, the emphasis is on oil and gas multiphase flow and applications in hydrocarbon recovery, as well as carbon dioxide storage in oilfields and aquifers.

Even though renewable energy technologies are developing fast, not least because ambitious efforts have been put to reduce CO₂ emissions because of the Paris Agreement, oil and gas are still and will play an important and irreplaceable role in energy system for decades to come (<https://www.iea.org/weo2018/oilandgas/>). According to the International Energy Agency, 53% of global primary energy demand in 2040 will still be provided by oil and gas even if there is a transition to a low-carbon economy, as shown in Figure 1.1. To take full advantage of the cost of oil/gas exploration, facilities and production wells, appropriate displacement methods and strategies should be identified based on a detailed physical description of multiphase flow behaviour in the subsurface. Thus, it is important to capture multiphase flow behaviour in porous media to help enhance oil recovery.

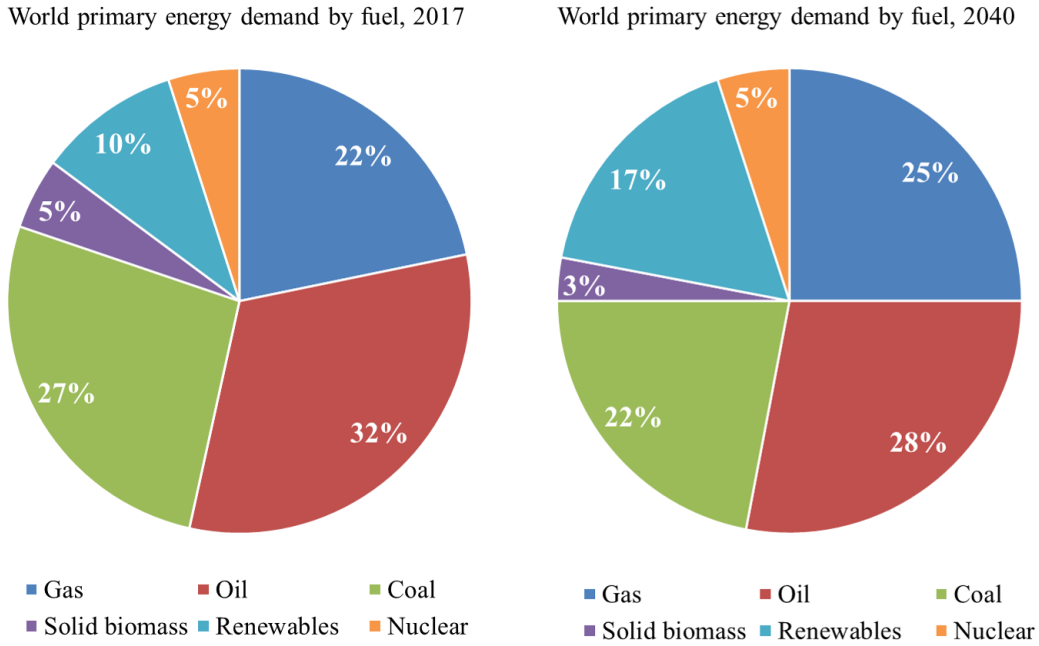


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1.1 Multiphase flow in subsurface rock

Multiphase flow in geological porous media exists in a wide range of subsurface applications ranging from natural science, such the hydrologic cycle, to the fields of energy, such as oil and gas production (Parker, 1989; Morrow and Buckley, 2011), as well as carbon dioxide storage (Bachu, 2003; Andrew et al., 2013). In particular, the petroleum industry is interested in multiphase flow, to remove oil as effectively as possible in the presence of other phases, water and possibly gas.

Most of our energy comes from oil and gas extracted from porous rock. To produce more oil and gas, hydrocarbons are displaced in the pore-space of geological media with water. While single-phase flow has been well characterized, many phenomena of steady-state immiscible two-phase flows – here oil and water – in porous media are still not adequately understood. The macroscopic flow of immiscible phases in porous rock is determined by complex microscopic processes. Fundamentally, multiphase flow is controlled by the interplay of viscous and capillary forces whose balance depends on the geometry and topology of the porous structure, wettability, and flow regime (Blunt, 2017). It is the behaviour at

the sub-millimetre scale of the pores within the rock that plays a key role in determining the overall movement of the fluids and how much can be recovered. In combination with the huge practical applications that drive the science, recent scientific advances have transformed our understanding of how fluids move at the pore scale: the development of methods to image rocks, the pore space and the fluids within them.

1.2 Scientific Achievements

In this thesis, we devised an experimental method based on differential imaging to examine how flow rate impacts the pore-scale distribution of fluids during co-injection in both homogeneous sandstone and heterogeneous carbonate at both low and high capillary numbers. The micro-CT imaging method and two-phase flow experiments were combined to visualize the fluid distribution of oil and brine and to develop a better understanding of flow behaviour at the pore scale. This enables us to elucidate flow regimes (connected, or breakup of the non-wetting phase pathways) for a range of fractional flows in disordered porous media using X-ray micro-tomography techniques.

To improve our understanding of the multiphase flow in porous media, fast synchrotron X-ray CT combined with sensitive pressure differential measurements were used to study the steady-state flow of equal volume fractions of oil and gas as a function of flow rate.

The main achievements of the research presented in this thesis are:

- 1) I developed an X-ray tomography-based experimental and image analysis methodology to measure the relative permeability and associated pore occupancy in Bentheimer sandstone at low and high capillary numbers.
- 2) During the hour-long scan time, an intermittent flow was identified by obtaining the differential image between a doped brine image and the scans taken at each fixed fractional flow. The pressure drops at each fractional flow at low and high Ca were measured by high-precision pressure differential sensors and used to calculate the relative permeability. The measured relative permeabilities compared well with literature measurements on larger cm-scale cores.
- 3) I expanded this method and applied it to a heterogeneous Estailades carbonate. Dif-

ferential imaging was applied to (a) distinguish micro-porous regions from macro-pores, and (b) determine fluid phase pore occupancy and relative permeability at high capillary number. We find that a fraction of the macroscopic pore space contains an intermittency in addition to brine and decane, which is occupied either by brine or oil during the hour-long scan time. Furthermore, fluid occupancy in micro-porosity was classified into three sub-phases: micro-pore space with oil, micro-pore space with brine, and micro pores partially filled with oil and brine. This allows us to assess for the first time the initial and recovered oil from micro- and macroporous regions.

- 4) Synchrotron X-ray micro-tomography imaging and experiment were combined to study the wetting and non-wetting phase flow behaviour as a function of flow rate. At low capillary numbers, the pressure gradient, ∇P , across the sample was stable and proportional to the flow rate Q (and hence capillary number). As the capillary number increased beyond $Ca \approx 10^{-6}$, pressure fluctuations were observed combined with local rearrangements of fluid in the pore space, leading to a small decrease in the non-wetting phase saturation, while retaining a linear dependence between flow rate and pressure gradient for the same fractional flow. Beyond a critical value $Ca > 10^{-5}$ we observed a power-law dependence with $\nabla P \sim q^a$ with $a \sim 0.6$ combined with rapid fluctuations of the pressure differential of a magnitude equal to the capillary pressure.

1.3 Thesis outline

The aim of this research is to provide comprehensive information on multiphase flow in porous media, and to develop novel methods to find relative permeability and pore occupancy at the pore scale.

This thesis is organised as follows:

Chapter 2 introduces a review of the literature relevant to this work and background information for multiphase flow in porous media and X-ray imaging.

Chapter 3 provides image acquisition methods, processing methods and analytical techniques that were applied in this work.

Chapter 4 describes the experimental methods used in this research including the appa-

ratus and imaging strategies.

Chapter 5 demonstrates the results of relative permeability measurement and pore occupancy evaluation in Bentheimer sandstone and Estailades carbonate for mm-scale experiments using micro-CT imaging methods.

Chapter 6 provides results from dynamic multiphase fluid flow experiments using synchrotron-based X-ray microtomography. We classified the flow regimes into three regions based on the fluid flow behaviour and pressure difference.

Chapter 7 presents the conclusions of this research and provides suggestions for future work including studies related to correlative imaging, dynamic imaging, and modelling of multiphase flow.

Chapter 2

Scientific background and literature review

Understanding multiphase flow in porous media is of vital importance in many subsurface applications. Pore-scale studies can explain the fluid flow mechanisms behind models conducted at the core (cm) scale and field (km) scales (Blunt et al., 2013). With the rapid development of imaging methods, pore-scale research has made great progress towards visualizing what happens within pores, providing insight and benchmark data for larger-scale modelling.

In this chapter, we will review basic concepts of fluid flow at the pore scale, and the controls on flow regimes, while providing a description and the characteristics of relative permeability. A brief introduction to pore-scale flow mechanisms is used to demonstrate the main forces controlling flow in porous media, followed by a presentation of the Darcy's law governing flow at the core scales. Displacement processes including imbibition and drainage are then introduced to describe the mobility of the fluids. After that, we illustrate the principles of mobilization of oil ganglia. We then discuss the flow regimes observed by X-ray imaging technology. A definition of the Euler characteristic, evaluating connectivity of the non-wetting phase, follows. At the end of this section, a summary is presented with a statement about how we will proceed to the next chapters.

2.1 Pore-scale flow mechanisms

Different forces acting on the fluids together control how fluids distribute in the pore space with complicated geometries and topologies during the simultaneous flow of more than one fluid phase in porous media (Blunt, 2017). Multiphase flow in porous media is governed by the interplay of viscous and capillary pressures. We will study the simultaneous flow of oil and water through permeable rock, in which interfacial tension, capillary pressure and viscous pressure are all considered. In this section, more information about these pressures, how they are defined, and factors that impact their characteristics will be described.

2.1.1 Interfacial tension

Interfacial tension (also known as the surface energy density) is the elastic tendency of a fluid surface which makes it acquire the least surface area possible. It is the energy per unit area of the surface between two phases, or the change in free energy for a change in area: $\sigma = dF/dA$. It arises from the fluid-fluid interface that helps to maintain the largest intermolecular bonding with itself and the lowest between two dissimilar fluids.

There are two main methods to measure interfacial tension in the laboratory: the capillary rise method and the pendant drop method (Adamson and Gast, 1967). The capillary rise method is the oldest method used for determining interfacial tension. A thin circular capillary is dipped into the tested liquid. If the interaction forces of the liquid with the capillary walls (adhesion) are stronger than those between the liquid molecules (cohesion), the liquid wets the walls of tubing and rises to a defined with a hemispherical meniscus (Richards and Carver, 1921). The second method can be used with a selected plane (Andreas et al., 1938) or with drop shape analysis (Butler and Bloom, 1966). The shape of a liquid drop (hanging from a surface) can be described by the Laplace-Young equation, which is a balance between gravity, hydrostatic pressure, and interfacial tension effects (Zeppieri et al., 2001). Liquid droplets (small gas bubbles in liquid) can change the shape from that of a sphere due to gravity, as Figure 2.1 shows. The interfacial tension can be calculated from the droplet or bubble shape, which is given by

$$\frac{1}{R} + \frac{\sin \phi}{x} = \frac{2}{R_0} - \frac{\Delta \rho g z}{\sigma} \quad (2.1)$$

where R is the radius of curvature at the point (x,z) , R_0 is the radius of curvature at the apex of the drop, g is the gravitational constant, $\Delta\rho$ is the effective density of the liquid drop, σ is the interfacial tension, ϕ is the angle made by the tangent at the point (x,z) and the x coordinate axis.

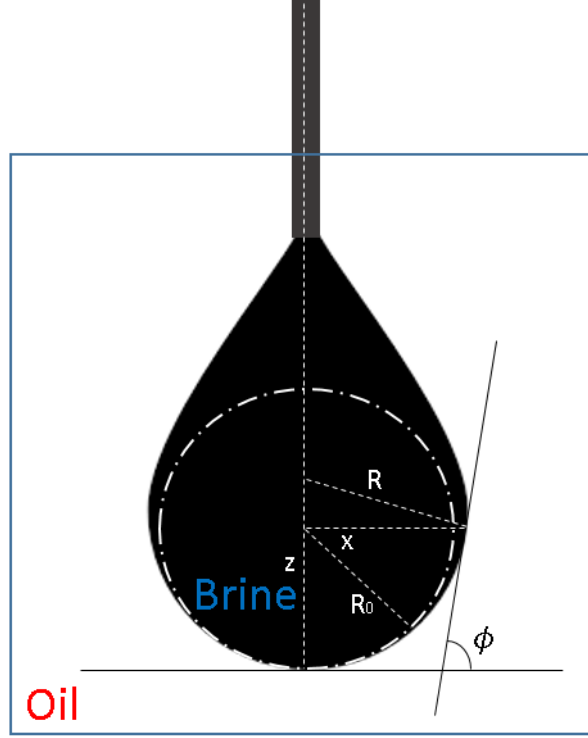


Figure 2.1: Pendant drop showing the geometrical variables in Equation (2.1).

2.1.2 Capillary pressure

The local capillary pressure is commonly defined as the difference in pressure between two phases at the pore scale, which depends on the curvature and interfacial tension at equilibrium as given by the Young-Laplace equation (Leverett, 1941; Morrow, 1970),

$$P_c = P_o - P_w = \sigma \left(\frac{1}{r_1} + \frac{1}{r_2} \right) \quad (2.2)$$

where P_c [Pa] is the capillary pressure, σ [N/m] is the interfacial tension between the two phases, P_o [Pa] is the pressure in the oil, P_w [Pa] is the pressure in the water phase, and r_1 [m] and r_2 [m] are the principal radii of the curvature of the interface.

If we consider a pore/throat that is a circular cylinder of radius r containing two phases with a contact angle θ , measured through the water, then capillary pressure can be defined as:

$$P_c = \frac{2\sigma}{r} \cos \theta \quad (2.3)$$

2.1.3 Viscous pressure

Fluid viscosity is a measure of its resistance to flow by shear stress or tensile stress, which is the property of a fluid which prevents the relative movement between two surfaces of the fluids that are moving at different velocities. In short, viscosity means the friction caused by the molecules of the fluid. Viscous pressures are proportional to the rate at which the fluid velocity is changing in space.

By rearranging Darcy's law, the viscous pressure is

$$\Delta P_{visc} = \frac{q\mu L}{k} \quad (2.4)$$

where q [m/s] is the Darcy velocity which is the fluid volume of flowing per unit cross sectional area of the porous medium per unit time, μ [Pa·s] is the fluid dynamic viscosity, L [m] is the length, k [m²] is the absolute permeability.

2.1.4 Capillary number

Capillary number is defined as the dimensionless ratio between the viscous pressure drop and capillary pressure, which can be used to distinguish flow behaviours.

For a pore of radius r and length L , the capillary force is given by the Young-Laplace equation (2.3), and the viscous forces by arranging Darcy's law (2.4). The ratio of viscous pressure difference to capillary pressure is then given by

$$\frac{\Delta P_{visc}}{P_c} = \frac{q\mu L}{k} \frac{r}{2\sigma \cos \theta} \frac{1}{\sigma} \propto \frac{Lr}{2k} \frac{q\mu}{\sigma} \quad (2.5)$$

which is the capillary number. This number describes the ratio of these effects on the pore

scale and is commonly expressed as

$$Ca = \frac{q\mu}{\sigma} \quad (2.6)$$

where q [m/s] is the Darcy velocity, μ [Pa·s] is the viscosity, and σ [N/m] is the interfacial tension.

A macroscopic capillary number (Armstrong et al., 2014) considering the length of clusters of oil at the pore scale can be written as:

$$Ca^{macro} = \frac{qL\mu_w}{kP_c} \quad (2.7)$$

where L [m] now represents a cluster length, k [m²] is the permeability, and P_c [Pa] is a capillary entry pressure. In this definition, viscous and capillary pressures are equal at $Ca \approx 1$. This definition is more intuitive compared with that obtained from Equation (2.6); however, the cluster length must be calculated which is not easy to obtain when the cluster connects the inlet to the outlet using pore scale imaging and so is not well defined.

2.2 Multiphase flow in porous media

Macroscopic single phase flow is described using Darcy's law in porous media:

$$q = -\frac{k}{\mu}(\nabla P - \rho g) \quad (2.8)$$

where q [m/s] is the Darcy velocity which is the volume of flowing fluid per unit cross sectional area of the porous medium per unit time, k [m²] is the absolute permeability which is a constant depending on the geometry of the pore space, μ [Pa·s] is the fluid dynamic viscosity, P [Pa] is the pressure, ρ [kg/m³] is the fluid density, g [m/s²] is the acceleration due to gravity.

When more than one fluid is present in the system, the permeability of each phase reduces because the flow of each phase is blocked by the presence of the other phases. In this case, multiphase flow in disordered porous media is summarized by an empirical extension,

$$q_i = -\frac{k k_{r,i}}{\mu_i}(\nabla P_i - \rho_i g) \quad (2.9)$$

by introducing relative permeability here to describe flow of each phase in the system, where $0 < k_{r,i} < 1$ is the relative permeability for phase i . Generally, the sum of relative permeabilities over all phases is less than 1.

Relative permeability is the fundamental concept for macroscopic two-phase flow in porous media. The relative permeability of each phase is a function of saturation for a given rock. It has great significance to the oil industry as relative permeability is a key input to reservoir simulation to guide and predict oil production. Relative permeability is used in fractional flow theory, which was first introduced by Leverett (1941) and Buckley and Leverett (1942). Fractional flow theory has been widely used in analytical immiscible multiphase flow studies (Osoba et al., 1951; Terwilliger et al., 1951; Rapoport and Leas, 1953; Jones-Parra and Calhoun Jr, 1953).

However, it is still poorly understood how exactly the two fluids are configured within the pore space, and how this in turn influences their continuum scale relative permeabilities. Oil and water relative permeabilities are assumed to be proportionality functions in Darcy's law, generalized to the case of multiphase flow (Bradford and Philip, 1970; Craig, 1971; Wooding and Morel-Seytoux, 1976; Adler and Brenner, 1988). Even though Darcy's law for one-phase flow has received wide validation for numerous types of porous media, further work is needed concerning most aspects of multiphase flow and the proper generalization of Darcy's law. The criticism towards the applicability of relative permeability in macroscopic models centres on whether relative permeabilities depend on the fluid flow rates and on the specific manner in which the fluids are distributed in the pore space (Leverett, 1941; Chatenever and Calhoun Jr, 1952; Jones and Roszelle, 1978; Avraam and Payatakes, 1995a,b; Rose, 1999; Tallakstad et al., 2009a).

2.2.1 Factors impacting relative permeability

Factors that impact relative permeabilities have been studied for many years: (a) saturation of each phase, which is the critical parameter (Johnson et al., 1959; Naar et al., 1962; Avraam and Payatakes, 1996); wettability (Spiteri et al., 2008; Khishvand et al., 2016; Zhao et al., 2016; Zou et al., 2018); viscosity ratio (Dias and Payatakes, 1986; Avraam and Payatakes, 1995a,b; Reynolds and Krevor, 2015); saturation history (Dana and Skoczylas, 2002; Alizadeh and Piri, 2014); and pore geometry and topology (Ott et al., 2015; Prodanović

et al., 2015).

2.2.1.1 Capillary number

Due to capillary pressure, flow is blocked in narrow throats or small pores at low capillary number. Increasing capillary number represents a relative increase in viscous pressures which forces oil (the non-wetting phase) through the pore space during water (the wetting phase) injection. As a consequence the relative permeability of the injected phase will increase at high Ca (Avraam and Payatakes, 1996; Reynolds and Krevor, 2015).

At the pore level, the balance between viscous pressure and capillary pressure impacts the local saturation distribution. For example, the residual oil saturation varies when flow rate changes (Abrams and Prausnitz, 1975). As introduced in Section 2.1.4, the capillary number $Ca = \mu q / \sigma$, is usually used to identify the relative magnitude of viscous to capillary pressures. At reservoir conditions, typical values for σ_{ow} are of the order of 10^{-3} N/m, and q is of the order of 10^{-6} m/s (Blunt, 2017). Generally, the displacement process is controlled by capillary pressures when $Ca < 10^{-6}$ (Niu et al., 2015), while viscous pressures are dominant when $Ca > 10^{-3}$ (Blunt, 2017). Most natural flows have Ca in the range 10^{-8} - 10^{-5} , although the flow close to wells and near miscible flow can have Ca as high as 10^{-3} (Blunt, 2017).

Darcy's law, Equation (2.9) suggests a linear relationship between flow rate and pressure gradient. However, Sinha and co-workers demonstrated that, in a glass bead pack experiment and associated simulations (Sinha and Hansen, 2012; Singh et al., 2017), there is no Darcy regime for capillary numbers lower than $10^{-4.75}$. They concluded that there is a power-law scaling of flow rate with pressure drop across the system with an exponent $a \approx 0.5$. When capillary numbers are $10^{-4.75}$ or higher, the pressure drop varies linearly with Ca . In order to describe capillary flow mechanisms and study further the relationship between relative permeability and capillary number, multiphase flow experiments combined with X-ray imaging techniques on the microscopic level will be applied to study the fluid behaviour at the pore scale in this thesis and to quantify the relative permeability through sensitive pressure measurements.

2.2.1.2 Rock heterogeneity

Heterogeneity means that macroscopic quantities tend to be different between samples of the same rock type. Carbonate samples often present heterogeneity at the core scale because of the presence of a secondary porous system such as sub-resolution pores and fractures, which makes it difficult for simulation and accurate assessment of reservoir performance. Since carbonate reservoirs hold more than half of oil and gas reserves, advanced approaches should be developed and improved to better understand the fluids and flow properties within these complex porous formations, including a detailed understanding of the fluid behaviour, relative permeabilities, and natural pore structure at various scales (Gouze et al., 2008; Gharbi and Blunt, 2012; Bijeljic et al., 2013a; Blunt et al., 2013; Ott et al., 2015; Apourvari and Arns, 2016; Gao et al., 2019). In particular, micro-porosity, pores of sub-micron size, may control the flow and transport behaviour, but is challenging to describe using current imaging and modelling methods (Ghous et al., 2007; Tian and Wang, 2018).

Micro-porosity in single-phase flow in carbonates (Bijeljic et al., 2018; Lin et al., 2016) and two-phase flow in a dual porosity sandstone (Lin et al., 2017) has been quantified using differential imaging micro-tomography. In this work we will extend these techniques to study two-phase flow in a micro-porous carbonate.

2.2.2 Measurement methods

Conventional measurements of relative permeabilities are calculated using Darcy’s law, as shown in Equation (2.9). It has been observed that the characteristics of relative permeability can exhibit a strong path dependency, or hysteresis, which means that the properties depend on the direction of saturation change and the displacement sequence.

Conventional laboratory relative permeability measurements are classified into steady-state and unsteady-state methods (Honarpour and Mahmood, 1988). In steady-state methods, two immiscible phases are injected simultaneously into a rock sample at controlled flow rates at a specified fractional flow until the system reaches equilibrium, which can be indicated by a stable pressure drop, and a constant fluid saturation profile along the core. The fractional flow is then altered in increments to reproduce a flooding sequence. The advantages of this method are that: (a) this technique can make sure capillary equilibrium has been achieved;

(b) the effective permeability of each phase can be easily calculated using Darcy’s law. The disadvantages are that: (a) this is a time-consuming process, which takes a very long time to reach equilibrium between two phases at each fractional flow (Honarpour and Mahmood, 1988); (b) the wetting phase may be retained at the end of the core, meaning that the saturation profile is not constant. This so-called capillary end effect has to be corrected (Osoba et al., 1951; Richardson et al., 1952; Lin et al., 2018).

In unsteady-state methods, the experiment is performed under dynamic displacement. Pressure differences and the corresponding saturation are monitored as a function of time (Johnson et al., 1959). The biggest advantage is that this method is much quicker than a steady-state experiment. Compared with the steady-state method, the disadvantages are: (a) capillary equilibrium is not reached; (b) the measurement of relative permeability is calculated using fractional theory instead of Darcy’s equation directly (Welge, 1952; Johnson et al., 1959; Jones and Roszelle, 1978); (c) uncertainty may be introduced because of viscous fingering and the combination of capillary and viscous effects (Dake, 2001).

In summary, the steady-state method is considered more representative of reservoir flow (Honarpour and Mahmood, 1988; Dake, 2001). In this study, we use the steady-state method to measure relative permeability.

2.3 Drainage and imbibition

The processes of drainage and imbibition depend upon wettability and the fluid originally occupying the pores. These play an important role on mobility change, describing fluids behaviours in real rock. Mobility is the fraction of total flow capacity for a particular phase. On the macroscopic level, both processes result in saturation changes.

2.3.1 Drainage process

Drainage is a fluid flow process that occurs when the non-wetting phase displaces the wetting-phase in a porous medium, which leads to an increase in the non-wetting phase saturation. During this process, the mobility of the non-wetting phase increases while that of the wetting phase decreases. For example, in a water-wet reservoir, gas/oil injection

represents a drainage process. The initial formation of the reservoir is a primary drainage process since oil or gas (non-wetting phase) enters a hydrocarbon reservoir.

Primary drainage occurs that when a non-wetting phase initially enters a porous medium filled with wetting phase, the non-wetting phase invades the widest pore space first and then progressively fill smaller pores. The non-wetting phase firstly invades the widest portions of the pore space and fill smaller regions progressively. For very slow displacements, the fluid configurations progress from one equilibrium arrangement to the next with the capillary pressure increasing (Blunt, 2017). There is a threshold capillary pressure at the narrow region of the pore space, which determines the sequence of filling and how much wetting phase is displaced at the microscopic level. Assuming the pore or throat is a circular cylinder of radius r , the capillary pressure for an interface residing across the pore is $P_c = 2\sigma \cos \theta_R / r$, as Figure 2.2 has shown. This threshold capillary pressure relates to the receding contact angle because of the movement of the interface between two phases.

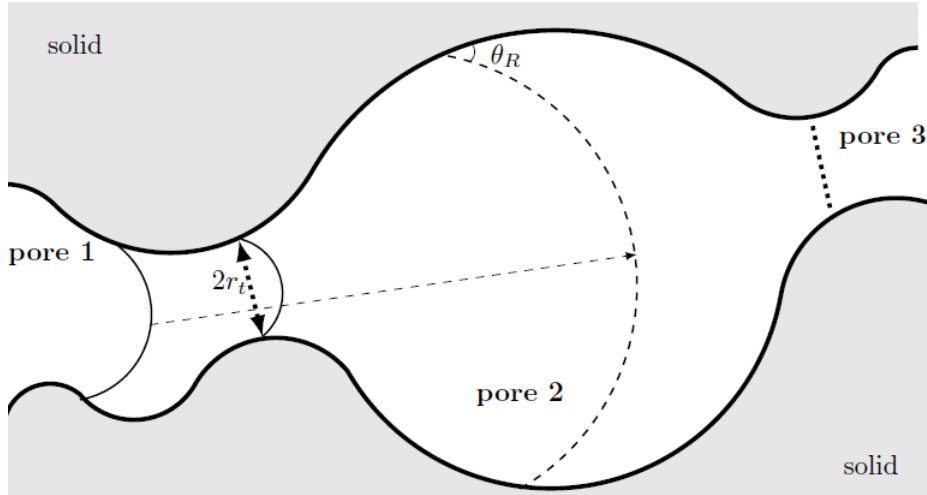


Figure 2.2: An illustration of the drainage process, where the wetting phase is displaced by the non-wetting phase. The terminal meniscus between the wetting phase and the non-wetting phase passes the throat and advances rapidly once the maximum capillary pressure is reached at the throat whose inscribed radius is r_t . Adapted from Blunt (2017).

2.3.2 Imbibition process

Imbibition occurs when the wetting phase displaces a non-wetting phase, leading to an increasing saturation of the wetting phase and a decreasing non-wetting phase saturation. As a result, the mobility of wetting phase increases as the wetting phase saturation increases.

Imbibition is common phenomenon happening in various fields. For instance, when the oil reservoir is water-wet, the process of waterflooding to produce oil is imbibition. It has an impact on areal and vertical sweep by advancing or hindering water movement.

Wettability determines which phase imbibes more. If a water-wet sample saturated with oil is put in water, water moves into the small pore spaces and oil is displaced. Then if an oil-wet sample saturated with water is placed in oil, oil can be imbibed into the sample.

As this study uses water wet samples, the following sections focus on behaviour in such samples.

2.3.2.1 Spontaneous imbibition

Generally, spontaneous imbibition describes the displacement process dominated by capillary pressures without any external pressure driving the phase into the rock (Schmid and Geiger, 2012; Mason and Morrow, 2013). During the process of pore filling, the wetting phase flows into the smallest pore spaces first and then progressively fill the other regions. The geometry of the pore and the sequences of throat-filling with wetting phase and how they are arranged all relates to the threshold capillary pressure during the pore filling process.

2.3.2.2 Snap-off and coalescence

Since oil (the non-wetting phase) occupies the centres of the pore space, the wetting-phase flows through the corners and the surface of the pore space and water layers form when the initial capillary pressure is high. When the water pressure increases, these water layers can swell and the water saturation increases.

When the wetting phase is unable to displace the non-wetting phase directly from an adjacent water-filled pore but the water layers continue to swell, snap-off occurs when the wetting phase fills the centre of the narrowest throat and pore. The breakup of the oil phase into ganglia is caused by snap-off events. Snap-off is a process where a ganglion is disconnected from the non-wetting phase (Blunt, 2017). Both pore geometry and wettability have an impact on snap-off (Wardlaw, 1982; Singh et al., 2017). Pickell et al. (1966)

studied the precondition of snap-off and concluded that snap-off only occurs when the radius of the growing oil blob is much larger than throat of the constriction in a water-wet rock. A snap-off event occurs when the capillary pressure is lower than the threshold pressure of the non-wetting phase (Lenormand et al., 1983). Generally, the non-wetting phase is always trapped in the larger pores and surrounded by the wetting phase. Dynamic filling procedures of snap-off during imbibition in a throat are shown in Figure 2.3 at the pore scale. The oil ganglion is trapped in the pore space after disconnecting through snap-off.

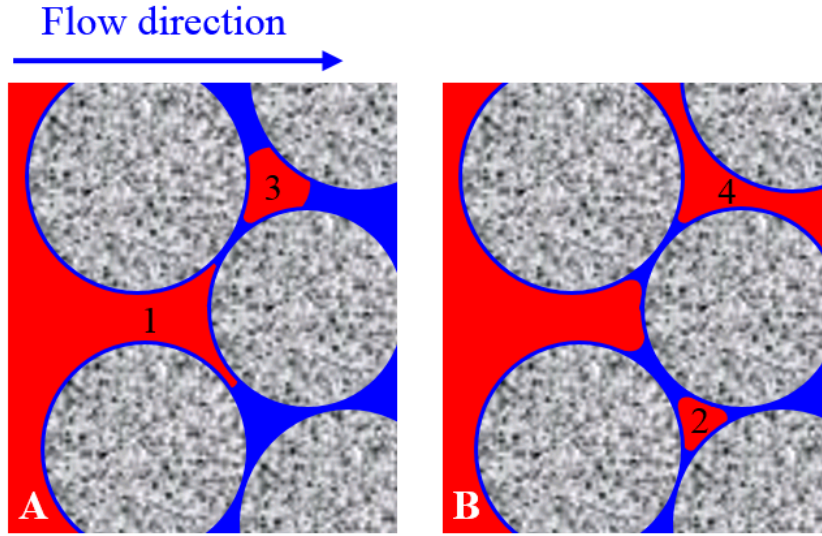


Figure 2.3: This figure shows a schematic of snap-off and coalescence. (A) The distribution of wetting phase and non-wetting phase. (B) The new distribution of the two phases because of fluid redistribution of snap-off and coalescence. One oil ganglion disconnects from the narrow part into ganglion 1 and ganglion 2, called snap-off. Ganglion 2 and 3 connects and becomes ganglion 4, called coalescence. Adapted from Rücker et al. (2015b).

Capillary trapping is related to the rock type and is controlled by fluid and interfacial physics at the size scale of rock pores (Andrew et al., 2014; Krevor et al., 2015).

A snap-off event can lead to other displacement processes, such as coalescence caused by the fluid redistribution after other snap-off events in the vicinity, shown in Figure 2.3(B) (Armstrong and Berg, 2013; Rücker et al., 2015b).

The distribution of ganglia sizes at residual saturation is controlled by pore-space structure and connectivity. Well-connected rocks tend to have more large ganglia relative to small ganglia and vice-versa. In the generalized theory for two-phase flow, breakup and coalescence are the processes responsible for the exchange between connected and disconnected phases (Doster and Hilfer, 2011).

2.4 Principles of oil movement

A ganglion is defined as a blob of the non-wetting phase (taken to be oil) that occupies at least one to several adjoining pores (Payatakes, 1982) and which is completely surrounded by the wetting phase. To understand oil recovery, we need to know how oil ganglia form, since, under capillary control, once the oil is surrounded by water it is usually assumed that it cannot move; and also to understand the conditions under which oil can still be displaced even if it, temporarily, resides in a ganglion.

2.4.1 Oil/water/rock interfaces

Several factors can impact the mobilization of a ganglion, including ganglion size, contact angle, capillary number and the geometry of the sample. When a ganglion partly touching the solid wall, the interfaces between the non-wetting phase and the wetting phase can always help to locate where the oil ganglion is and where the oil ganglion moves.

The equilibrium of static oil-water interfaces can be explained by the Young-Laplace equation (2.2). For a single interface/meniscus, the relationship of hydrostatic equilibrium is using both the Young-Laplace and Young equations:

$$P_c = P_o - P_w = \sigma_{ow}\kappa \quad (2.10)$$

$$\sigma_{os} = \sigma_{ws} + \sigma_{ow} \cos \theta \quad (2.11)$$

where P_c is the capillary pressure, P_o is the pressure of oil phase, P_w is the pressure of water phase, κ is the total curvature of the interface, σ_{os} is the interfacial tension between the oil phase and the solid phase, σ_{ws} is the interfacial tension between the water phase and the solid phase, σ_{ow} is the interfacial tension between the oil phase and the water phase and θ is the contact angle, which is shown in Figure 2.4. An oil ganglion may be surrounded by several interfaces if it resides in irregular pores and throats; the ganglion can only be stable if $\nabla P = 0$, which means that the pressure around the ganglion (water phase) is uniform as well as the pressure inside the ganglion (oil phase). In this condition, the capillary pressure at each orientation is the same across the interface.

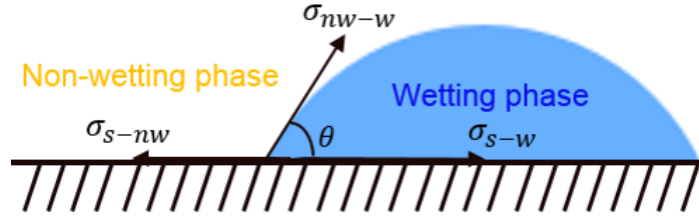


Figure 2.4: Fluid arrangement satisfying Young's equation.

2.4.2 Mobilization of oil ganglia

When there is a flow and the viscous pressure between oil-water interfaces is larger than the capillary pressure, the oil ganglia start to mobilize. In this condition, the contact angle depends on the direction of movement. The advancing contact angle, where water displaces oil, is larger than the receding contact angle that recedes towards water phase, shown in Figure 2.5. When an external pressure gradient is applied, oil ganglia move when the

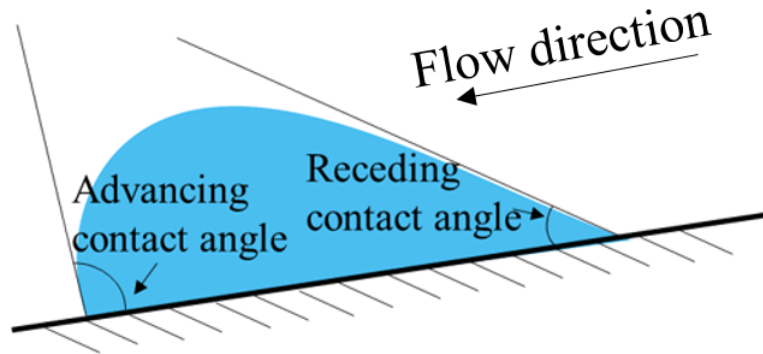


Figure 2.5: Hysteresis of the contact angle in two-phase flow. The arrows give the direction in which the interface of two-phase flow moves. Water is shown in blue.

downstream interfaces reach a critical drainage curvature, which is related to the geometry of the pore/throat and the limiting receding contact angle (Payatakes, 1982), as Figure 2.6 shows. When the system is at equilibrium, the oil ganglion stays as Figure 2.6(A) shows. When the wetting phase is injected from the left side, there is a pressure gradient in the system. Before mobilization, the shape of the ganglion changes, Figure 2.6(B), and the curvature at the left side becomes smaller. Then when the pressure gradient is larger than the critical drainage curvature, the ganglion starts to move as shown in Figure 2.6(C).

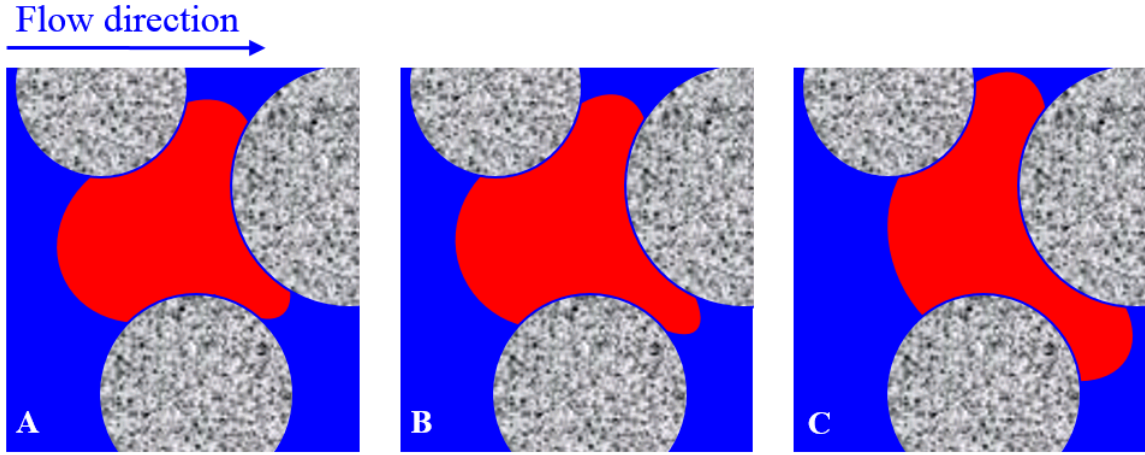


Figure 2.6: Mobilization process of an oil ganglion. (A) The status showing that the ganglion is at equilibrium. (B) The ganglion before mobilization. Wetting phase injected from the left side and the geometry of the ganglion changes. As the pressure gradient is not large enough, the ganglion does not move. (C) When the pressure gradient is larger than the critical value for mobilization, the ganglion starts to move. Adapted from Payatakes (1982).

2.4.3 Flow behaviours

An oil ganglion which occupies several individual pores may undergo a sequence of breakup and coalescence processes, which contributes to mass transfer. There are several factors that impact this flow, including capillary number, pore geometry and topology, flow-rate ratio, and wettability. To understand and explain how fluids breakup during two-phase flow, direct imaging studies in the pore space are useful.

One of the first observations of channel flow and slug flow were first observed in flow cells made with a single layer of glass beads (Chatenever and Calhoun Jr, 1952). When oil and water were flowing at less than 3×10^{-3} m/s, both fluids flowed through their own network of stable pathways, which was named as the channel flow regime. However, when the flow rate was increased to more than 6×10^{-3} m/s, slug flow was seen as the non-wetting phase was disconnected inside a continuous pathway of the wetting phase.

More flow regimes were observed and defined by Avraam and Payatakes (1995b, 1996). Their experiment was conducted in a planar model pore network of the chamber-and-throat type which was etched in glass and filled with oil and water. They combined optical observations and macroscopic measurements to determine flow regimes as well as the corresponding relative permeabilities. Four flow regimes were recorded and named based on the

size of mobilized ganglia, the fraction of ganglia, and the presence or absence of connected pathways: large-ganglion dynamics (LGD), small-ganglion dynamics (SGD), drop-traffic flow (DTF), as well as connected pathway flow (CPF), as shown in Figure 2.7.

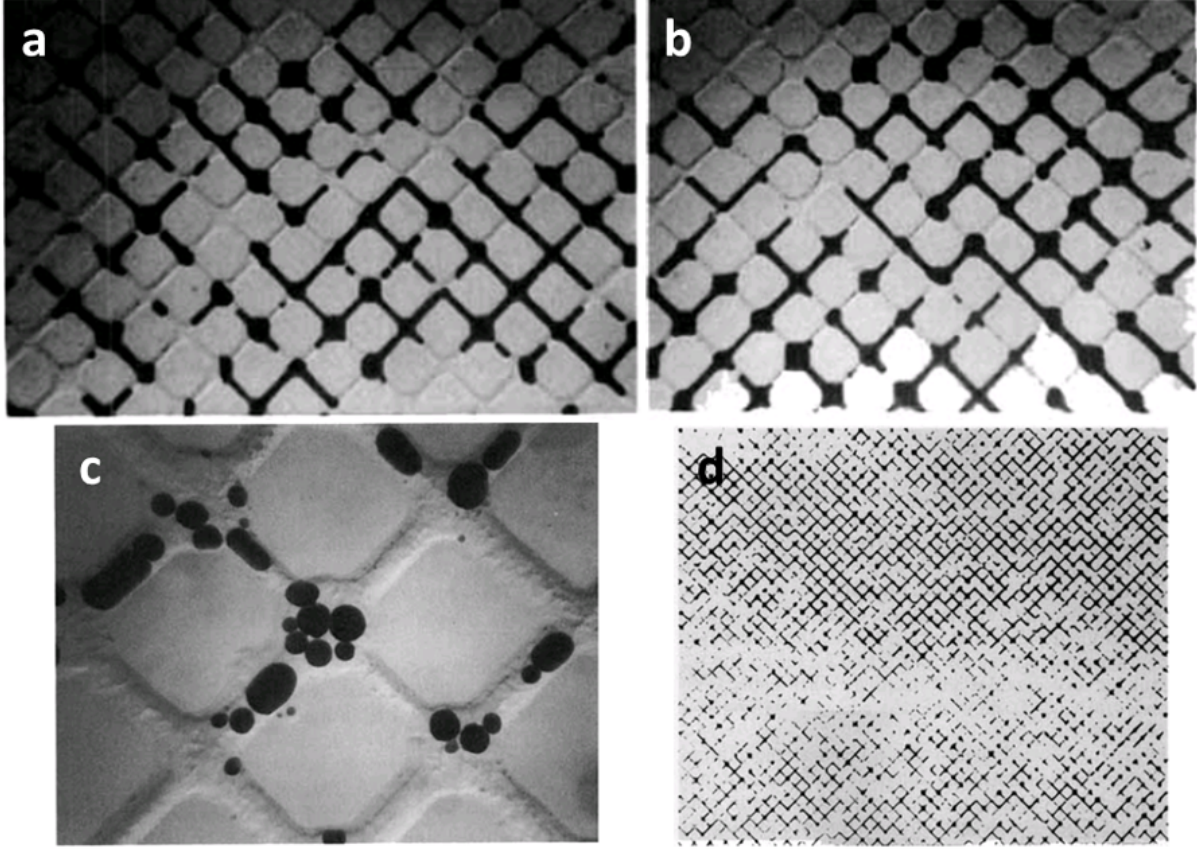


Figure 2.7: Images of the four flow regimes in micromodels, modified from Avraam and Payatakes (1995b). (a) Large-ganglion dynamics. (b) Small-ganglion dynamics. (c) Drop-traffic flow. (d) Connected pathway flow.

Tallakstad et al. (2009a,b) conducted an immiscible air (low viscosity) and water (high viscosity) flow experiment in a quasi-two-dimensional porous medium at steady state and recorded this using a digital camera. Oil fragmentation and mixing were observed at high capillary number. The pressure drop was recorded for the whole experiment and they observed that there is a power-law relationship between the pressure gradient and the capillary number. Also, they found that the non-wetting phase saturation decreases when Ca increases.

Because flow in three-dimensional model might be different from two-dimensional models, as well as the opacity of the fluids, three-dimensional models and advanced imaging facilities, such as advanced microscopy, were required to capture two-phase flow in more realistic

systems at pore-scale resolution. Confocal microscopy techniques have been used to visualize flow in a 3D bead pack (Krummel et al., 2013). They demonstrated that the ganglia configurations are closely related to flow history: the typical ganglion size, and the total amount of residual oil, decrease as Ca increases. Oil is snapped-off in narrow throats during imbibition because the wetting phase flow through the layers coating the solid phases. When the capillary number increases during drainage, the typical ganglion size and the residual oil saturation decrease.

A 3D bead pack has been used to explore oil and water flow behaviour using confocal microscopy (Datta et al., 2014a,b). They compared flow behaviours at different flow rates and fluid viscosities. They observed that when two phases were injected simultaneously, when the sum of viscous pressures exceeds the capillary pressures at the pore scale, the non-wetting phase can break up into discrete ganglia at sufficiently large flow rates (high capillary numbers) while both phases flow in connected pathways for small flow rates (low capillary numbers). Through the competition between viscous and capillary pressures, the transition from fully connected to broken up flow was summarized.

As real rocks has complex pore structures and rough surface, flow in porous models can not represent that in real rocks. Furthermore, it is hard to see fluids flow inside real rocks with confocal microscopy. To solve these problems, X-ray imaging techniques are introduced in next section.

2.4.4 X-ray imaging applications

X-ray imaging is a powerful non-destructive technique that provides reliable three-dimensional images of opaque materials. Many authors have studied pore structure and fluid flow using this technique (Bijeljic et al., 2013a; Wildenschild and Sheppard, 2013; Akbarabadi and Piri, 2015; Menke et al., 2016; Al-Khulaifi et al., 2017; Alhammadi et al., 2017; Zou et al., 2018; Saif et al., 2019). Indeed, the use of rock imaging and analysis has become a routine procedure when studying multiphase flow in porous media (Blunt et al., 2013).

The trapped non-wetting phase in Silurian dolomite with complex structure, for which the pore size ranging from $0.2 \mu\text{m}$ to $20 \mu\text{m}$, was visualized and quantified at both low and high capillary numbers by Pak et al. (2015). Using X-ray micro-tomography imaging, the

effect of capillary desaturation on the cluster size distribution of the trapped non-wetting phase was analyzed. They demonstrated the occurrence of droplet fragmentation at high capillary number, and that this played an important role in oil production beyond capillary trapping. They concluded that drop fragmentation (cluster breakup) occurred mainly in pores with large aspect ratios (pore to throat ratio).

High-speed synchrotron-based X-ray imaging can capture real-time pore-scale events in porous media, which provides an efficient way to explore more fundamental and detailed physical phenomenon, such as pore-filling, snap-off, coalescence and ganglia dynamics during drainage or imbibition. For instance, Berg et al. (2013) observed Haines jumps and snap-off using this state-of-the-art technique. Singh et al. (2017) visualized and quantified how the amount and the rate of swelling of the wetting-phase layer impact snap-off during imbibition at reservoir conditions. Flow regimes from connected pathway flow to ganglia dynamics were identified and categorized in the fractional flow vs. capillary number space at the pore scale (Berg et al., 2015, 2016; Rücker et al., 2015a). Berg et al. (2015) captured the onset of oil ganglia mobilization during imbibition in a sandstone sample.

In the context of flow regimes, ganglion dynamics have also been observed using fast microtomography in a sintered glass bead pack, as mentioned above (Rücker et al., 2015a). In consolidated sandstone, it has been shown that imbibition in porous rock is a two-stage process that involves a combination of connected pathway flow and ganglion dynamics (Rücker et al., 2015b; Berg et al., 2016).

Reynolds et al. (2017) studied the flow of nitrogen and brine flowing at the same rate in Bentheimer sandstone: they demonstrated a dynamic connectivity of the phases where pathways of the nitrogen periodically connect and disconnect. This is a different type of behaviour, where ganglia of the non-wetting phase do not advect through the pore space, but instead local rearrangements of the pore occupancy allow flow sporadically, analogous to the stop-and-start of cars on roads controlled by traffic lights. Connected paths of the non-wetting phase are only seen at capillary numbers around 10^{-7} or lower, which is two orders of magnitude lower than the onset of ganglion dynamics observed in bead packs by Datta et al. (2014a).

There are thus two possible scenarios where a non-wetting phase is mobile even though not permanently connected between inlet and outlet. Firstly, viscous mobilization by the

externally applied flow field of the non-wetting phase pushes the non-wetting phase through the pore space. This occurs when the viscous pressure drop over a ganglion exceeds the capillary entry pressure of the next pore throat (see, for instance, Armstrong et al., 2014). In this case there is no driving force for viscous mobilization, regardless of flow rate. Secondly, there can occur an intrinsic mobilization triggered by a sequence of breakup and coalescence caused by local pressure fluctuations (Rücker et al., 2015a) referred to as dynamic connectivity by Reynolds et al. (2017). This behaviour can be captured using numerical simulation of multiphase flow through pore-space images. For instance, Armstrong et al. (2016, 2017) studied the flux contribution of disconnected phases as a function of fractional flow and capillary number. They demonstrated that at low non-wetting phase saturation, the dominant contribution to flow of the non-wetting phase was from disconnected phases over a wide range of capillary numbers.

2.5 Euler characteristic

The Euler characteristic is a number that can be used to characterise the topology of the ganglia of the different phases within the pore space. It is a topological invariant (Wildenschild and Sheppard, 2013; Herring et al., 2013), which remains unchanged for an object regardless of the way it is deformed. It can be determined from the Betti numbers (Wildenschild and Sheppard, 2013; Herring et al., 2013; Blunt, 2017):

$$Eu_{nw} = \beta_0 - \beta_1 + \beta_2 \quad (2.12)$$

where the subscript *nw* refers to the non-wetting phase, and β represents a Betti number (Vogel, 2002; Wildenschild and Sheppard, 2013; Blunt, 2017). In this study, β_0 represents the number of oil ganglia without holes (such as droplets within a single pore), β_1 represents the number of the redundant loops through pore throats within an individual ganglion, and β_2 is the number of isolated regions of water phase completely surrounded by oil.

Because the rock samples used are strongly water-wet the water is connected in layers everywhere. Results using the Euler characteristic are presented in Section 5.2.5.

2.6 Summary

In this chapter, the scientific background and literature review related to the topics in this thesis are presented. There are four shortcomings evident from the literature review which will be investigated in this thesis:

- 1) The most widely used experimental facility to improve our understanding of two-phase fluid rearrangements is synchrotron imaging. This takes an image within seconds but it is not always accessible, meaning that, to date, only a limited range of flow conditions have been studied at one fractional flow. Hence, it would be highly beneficial if lab micro-CT scanners can be deployed for this purpose since they are readily available, despite longer times required to acquire images. We will show how to measure steady-state relative permeability for a range of fractional flows and quantify intermittency using laboratory micro-CT scanning.
- 2) To date micro-CT has been widely used to study oil recovery in the larger macro-porous space that can be directly imaged; however, oil recovery in micro-porosity has not been studied so widely, as the micro-pores cannot be imaged explicitly using standard instruments. Ignoring micro-porosity can lead to very poor estimates of pore volume and saturation, especially for carbonates with complex pore structure. Carbonates are highly important, since they hold more than a half of hydrocarbon reserves in the world (Ahlbrandt et al., 2005). An important question is how to evaluate oil recovery from micro-porous carbonates using laboratory-based micro-CT? This will be addressed in the thesis using differential imaging.
- 3) Although steady-state oil-water flow behaviour has been studied for a range of capillary numbers in a homogeneous bead pack using confocal microscopy, it is not clear how the impact of viscous forces will be manifested in simultaneous flow of oil and water in real rock geometries, such as sandstones and carbonates. Thus, in this thesis we will seek answers to the following questions using rock samples: do oil and brine always flow in fixed connected pathways or is there a regime characterised by intermittent break up of the phase(s)? Which capillary number indicates the transition from connected fluid pathways to ganglia flow? How exactly do the oil clusters connect and disconnect, even during steady-state flow?

- 4) A non-linear scaling of flow rate with pressure gradient has been observed in a recent study for a limited range of capillary numbers from $10^{-5.4}$ to 10^{-4} in a bead pack (Sinha and Hansen, 2012; Singh et al., 2017). However, does the same relationship apply for a wider and lower range of capillary numbers from 10^{-7} to 10^{-5} in realistic rocks, covering capillary numbers representative of subsurface flow conditions and those near the wellbore? To investigate this, pressure measurements will be conducted for a range of capillary numbers from 10^{-7} to 10^{-5} and related to flowrates.

The development of X-ray computed micro-tomography techniques and two-phase flow experiment provide an effective way to observe flow regimes of three-dimensional porous media models and real rock samples. Specifically, the new contributions of this thesis, based on the discussion, are as follows:

- 1) To elucidate and quantify flow regimes as a function of capillary number.
- 2) To combine pore-scale imaging with steady-state relative permeability measurements, so that the relative permeability can be related to the pore-scale distribution of fluid.
- 3) To use differential imaging to quantify multiphase flow behavior in a micro-porous carbonate.

Chapter 3

Image acquisition and analysis techniques

This chapter explains the image acquisition and analysis methods employed in this thesis.

3.1 X-ray imaging technology

3.1.1 Attenuation theory

Generally, a three-dimensional digital image of rock sample is produced by the steps shown in Figure 3.1. When X-rays pass through the object, the sample image is enlarged through geometric magnification and converted to visible light through the scintillator camera, which is then optically magnified. This process results in one two-dimensional image. A series of 2D projection images at different angles are captured as the sample rotates 360° and then are reconstructed using a computer program to generate a 3D model.

During this process, as the X-ray beam passes through matter, the beam loses intensity. Its attenuation is described by the Lambert-Beer law. For a pure monochromatic beam, where all the photons have the same energy and wavelength, the attenuation of beam intensity

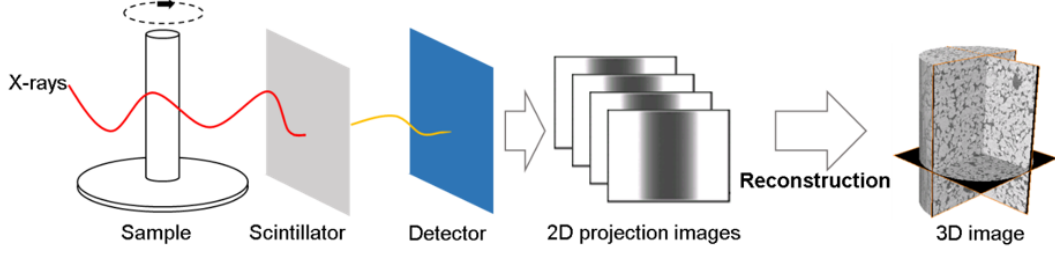


Figure 3.1: Schematic illustration of X-ray CT acquisition and reconstruction processes (adapted from Landis and Keane (2010)). A series of X-ray projection images are acquired and mathematically reconstructed to produce a 3D map of X-ray absorption in the volume. The 3D map is typically presented as a series of 2D slice images.

through a homogeneous object can be described as:

$$I = I_0 \exp(-\mu x) \quad (3.1)$$

where I is the attenuated intensity after the X-rays passing through the solid object of thickness x , I_0 is the original X-ray intensity, and μ is the linear attenuation coefficient. X-rays penetrate materials with a low attenuation coefficient more easily, while only a relatively short distance can be penetrated through a material with a high attenuation coefficient.

When the object is composed of multiple materials, the decay law becomes:

$$I = I_0 \exp\left(\sum_{i=0} -\mu_i x_i\right) \quad (3.2)$$

where μ_i represents attenuation coefficient of a single material i , and x_i is the thickness of this single material i . As the attenuation coefficients of various materials add linearly, the average linear attenuation coefficient can be estimated by considering volume fractions of each material. To identify each mineral in a multi-composite sample, many 2D images have to be taken at different angles to obtain the full 3D distribution of attenuation coefficients (Wildenschild and Sheppard, 2013).

3.1.2 Lab-based micro-CT

The experiments in this study were conducted in a Zeiss Versa XRM-500 and a Zeiss Versa XRM-510 micro-CT scanner.

The Zeiss Versa XRM-500 μ -CT scanner consists of a sealed X-ray transmission source, sample stage and contrast-optimized detector cameras. The tube voltage range of the X-ray source is from 30 to 160 keV with the maximum output 10 W. The detector system with detector turret consists of three objectives at different magnifications with optimized scintillators for highest contrast, including 0.4X, 4X and 20X. It can provide images consisting of 1000^3 - 2000^3 voxels. More details of the parameters used for this study will be described in Section 5.2.

This is a cone-beam micro-CT. The conical X-ray beam provides geometrical magnification by the position between X-ray source and detector, as shown in Figure 3.2. Firstly, sample images are enlarged through geometric magnification. Secondly, the scintillator converts X-rays to visible light and then the images are optically magnified by microscope optics before acquisition by a CCD detector. Here the X-rays are polychromatic and the beam is not collimated. The sample is rotated and the absorption of X-rays at different angles is recorded and used to produce a three-dimensional representation of the rock and fluids.

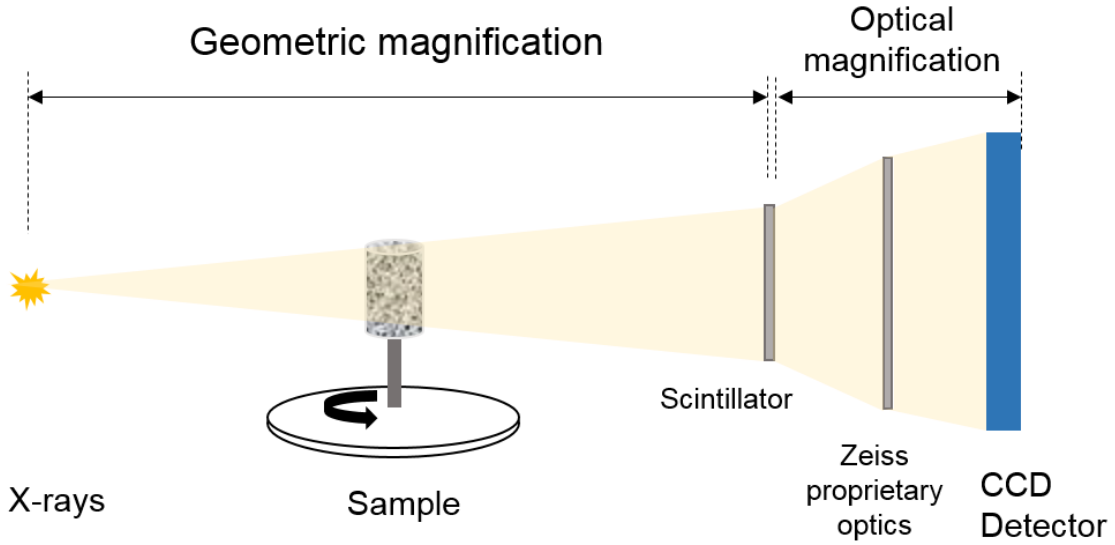


Figure 3.2: Zeiss XRM two-stage magnification architecture with a conical X-ray beam. The first stage is geometric magnification and the second stage is optical magnification.

Geometric magnification is defined as:

$$\text{Geometric magnification} = \frac{\text{Source to Detector Distance}}{\text{Source to Sample Distance}} \quad (3.3)$$

Total magnification, or pixel size of image resolution, is defined as:

$$\text{Total magnification} = \text{Optical magnification} \times \text{Geometric magnification} \quad (3.4)$$

A Zeiss Versa XRM-510 micro-CT was used in part of the study. A flat panel extension (FPX) was used in these experiments to extend the size of the field of view and shorten the scan time while improving image quality. A big advantage is that the size of the image that FPX can provide is as large as $3000 \times 3000 \times 2000$ voxels while that taken from the CCD detector is only $1000^3 - 2000^3$. Although both the CCD detector and the FPX can be used to study intermittency, images with the FPX took less time and had better quality. As Figure 3.3 shows, images are enlarged through geometric magnification and the flat panel design allows larger samples to be studied simply because the detector is larger. As Equation 3.3 shows, geometric magnification depends on the ratio of the source to detector and source to sample distance. Generally, we set the sample and source as close as possible to improve the magnification and also to decrease the scan time since the sample receives a larger X-ray flux. The distance between the sample and detector is decided by the voxel size of the image. Compared the images taken at the same voxel size from the CCD detector, the FPX can provide a larger image ($3000 \times 3000 \times 2000$) with better quality image within the same period of time. More details of the parameters set for this study will be described in Section 5.3.

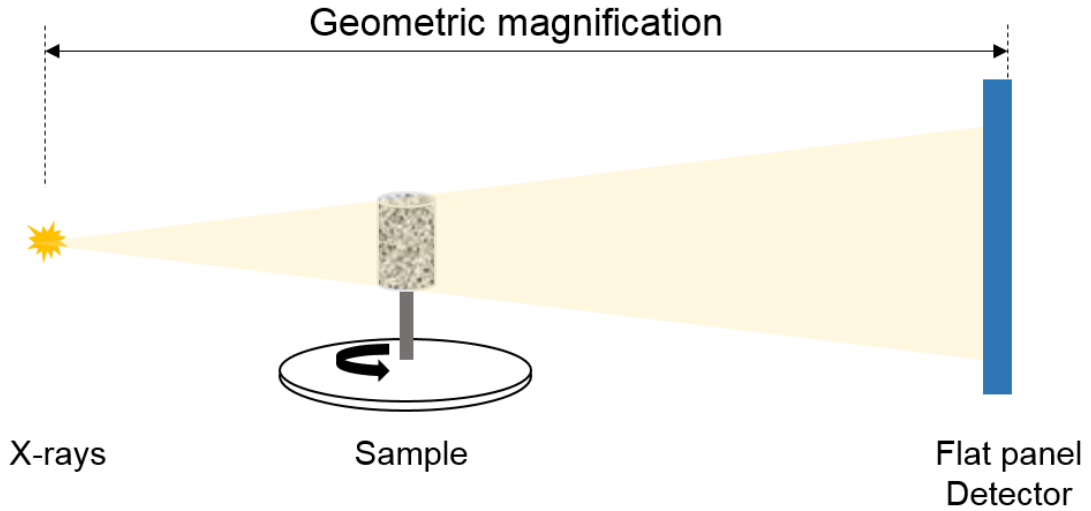


Figure 3.3: Zeiss XRM magnification architecture of flat panel with a conical X-ray beam.

3.1.3 Synchrotron-based micro-CT

For lab-based micro-CT, there is less constraint on the time when taking images. More than one hour is spent to take an image with good quality. However, flow and transport processes at the pore scale occur over a very short timescale, typically much less than hours. This implies that experiments are limited to studying static flow end points, or after a displacement process have finished. A synchrotron source offers a great advantage in that it provides a higher time resolution (order of 10s) than lab-based instruments.

A typical synchrotron consists of the electron gun, linear accelerator (Linac), booster synchrotron, storage ring, beam lines and the end station for different experimental applications, as shown in Figure 3.4. As a circular particle accelerator, a guiding magnetic field in

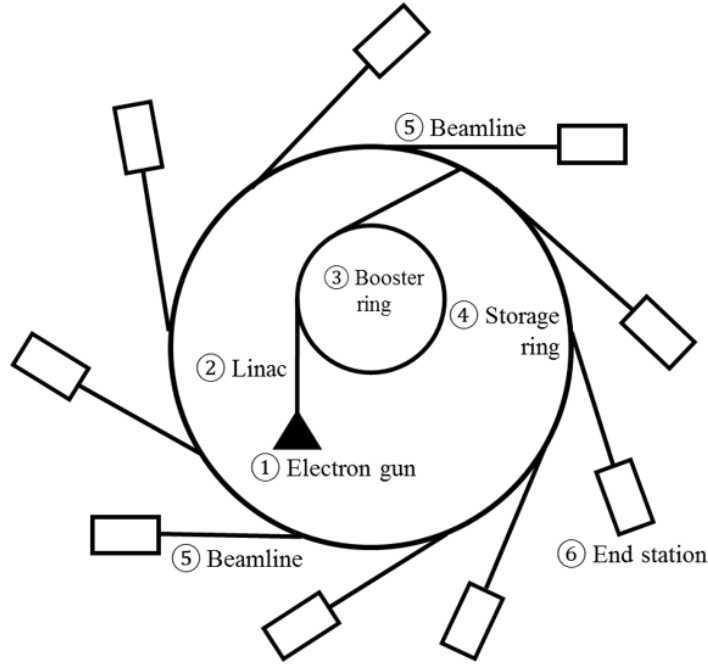


Figure 3.4: A schematic of the compositions of a synchrotron: ① Electron gun; ② Linac (the linear accelerator); ③ Booster ring; ④ Storage ring; ⑤ Beamline; ⑥ End station. Adapted from Menke (2015).

the synchrotron is used to accelerate, direct, focus and synchronize charged particles into a particle beam of increasing kinetic energy (Winick, 1995; Chao et al., 2013). Electrons, which are produced by an electron gun, are fired into a magnetic booster ring, in which the electrons are linearly accelerated through linac (a linear acceleration) to very high speeds, then the accelerated electrons are stored in a larger storage ring, which is maintained under vacuum conditions to minimize electron scattering off air molecules. Bending magnets and

insertion devices are the two sources for beamlines. Insertion device beamlines are used in this study.

Figure 3.5 shows the imaging schematic at the I13 beamline of Diamond Light Source. The electron beam is firstly accelerated by passing through the undulator following a wiggling or undulating path and emit X-rays. In our case, images was taken using photon energies ranging from 0 to 30 keV (Andrew et al., 2015). To discriminate adsorption in rock and fluids, mirrors are used to absorb the high-energy spectrum and filters are used to absorb the lower energies to narrow the spectrum to the desired band of radiation. Then the resultant X-ray beam passes through the rock sample, and the transmitted X-rays are collected by the detector. Here the detector is similar to the lab-based micro-CT mentioned in Section 3.1.2 but the scintillator must bear much higher energy fluxes, and the camera has to take and transmit images at high rates.

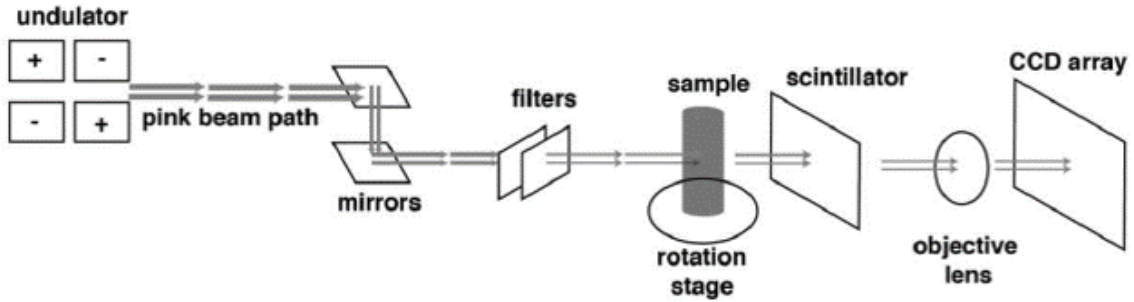


Figure 3.5: Schematic diagram of the pink beam synchrotron-based micro-CT apparatus (Menke et al., 2017). The beam passing through the undulator produces intense light. The spectrum is narrowed by mirrors and filters and then is transmitted through the sample. The scintillator fluoresces in the visible spectrum and the light is focused and measured by the objective lens and CCD camera array.

Instead of a conical source in lab-based micro-CT, a synchrotron X-ray beam is almost parallel with a stronger intensity. Because the X-ray beam is almost parallel at synchrotron sources, it is impossible to achieve geometrical magnification without X-ray optics with a high X-ray flux. If this is achieved, this high flux can be detected by a scintillator screen, converting the X-rays to visible light. In this way, high resolution is accessible by allowing the use of optical magnification lenses (Koch et al., 1998; Cnudde and Boone, 2013). This facility was used to capture dynamic flow behaviour and the results are shown in Chapter 6.

3.2 Image processing method

Before data analysis, 2D projections from micro-CT scanning at varying angles from -180° to 180° are reconstructed into a three-dimensional image, followed by filtering, then segmentation is completed to separate the fluid and solid phases.

3.2.1 Artefact removal

After acquiring a series of 2D projection images, these projections can be reconstructed into a three-dimensional volume dataset using a proprietary software associated with the micro-CT scanner. However, there are several artefacts that should be removed before further analysis. Generally, there are two artefacts: beam hardening and ring artefacts.

When a polychromatic beam passes through an attenuating material, lower energy photons are filtered out by the outer volume of the sample, which means that lower energy is easily attenuated in a beam spectrum and the average energy level of the scan increases (Ketcham and Carlson, 2001; Stauber and Müller, 2008). The beam is said to have become harder, and it attenuates less when it continues further into the material, which is called beam hardening (Wildenschild and Sheppard, 2013). This is one of the most common artefacts caused during micro-CT imaging. If the sample is homogeneous, this artefact causes the edges of the images to be brighter than the centre and this makes segmentation difficult because of a varying grey-scale distribution of one phase in one image. To avoid this beam hardening effect, one way is to use filters (copper, aluminium) between the source and sample to remove the low energy X-rays before they reach the sample. The other way is to use various algorithms during reconstruction to correct the beam hardening effect by assuming they are radially symmetric Gaussian functions and fitting a trigonometric or quadratic function to correct the intensity bias (Schlüter et al., 2014). In this study, the second method was applied.

Ring artefacts are caused by the defects in the detector array of a micro-CT, which may cause voxel intensity changes at the detector. These are observed as partial or full circles in the centre of the image. These can be removed in two ways: one is to image the background by moving the sample acquisition before scanning and then realigning the projections during reconstruction; the other is renormalizing the reconstructed image at the sinogram stage

using Fourier filtering (Münch et al., 2009). In this study, the image of background was taken pre-imaging. During the reconstruction, these projections are realigned.

3.2.2 Image registration and resampling

To explore flow regimes at different conditions in the same pore and throat structure, the same orientation must be assigned for all the images taken in one experiment. Even if the same position values in x , y and z direction are set fixed in the micro-CT scanner, small movements of the sample and imaging equipment cannot be avoided. Images taken at different conditions are registered to a reference image, such as the dry scan. After assigning the two images to similar positions manually, registration is applied by weighting the mutual information by similarity using the closest values to align the images (Li, 1990).

After this, images should be resampled to the same grid as the reference image. The voxels are translated, rotated and scaled in the same way as the reference image during this process. The most common methods for transforming the resample image are the standard and Lanczos methods. The standard method simply assigns the grey-scale value of each voxel in the registered images by using algebraic interpolation between adjacent voxels, but it loses vital image information. The Lanczos method applies a multivariate interpolation to the registered image by mapping each sample voxel to a translated and scaled copy of the Lanczos kernel which is evaluated at the new grid point. The sum of these translated and scaled kernels is then evaluated at the desired points (Lanczos, 1950; Burger and Burge, 2016). Lanczos method is slow but more accurate method that tries to approximate a low-pass filter that is in accordance with the sampling theorem. Thus the Lanczos method was used in this study.

3.2.3 Filtering

Noise is inevitable during the imaging. Filtering is the most effective way to reduce the noise from raw images and enhance the contrast between different phases.

The simplest filtering methods are mean and Gaussian filters. The main principle of the mean filter is to replace each voxel's grey-scale value with the mean value of its neighbours

(Ataman et al., 1981). The Gaussian filter method replaces each voxel's grey-scale value with the Gaussian weighted mean. Both methods work well, but the phase boundaries may become blurred, which is not suitable for later segmentation.

The median filter is the simplest edge-preserving filter method, and its principle is to replace the grey-scale voxel value with the median value of the local neighbourhood. Since the grey-scale value of the filtered image is related to the original voxel, the edge is preserved with conservative local groups. However, the edge-preserving ability is reduced when the amount of denoising is high (Andrew, 2014).

The anisotropic diffusion filter is another edge-preserving method, which smooths images without removing significant edges and lines that are important for the analysis and interpretation of the image (Perona and Malik, 1990; Catté et al., 1992). Anisotropic diffusion is a generalization of this diffusion process: it produces a family of parameterized images, but each resultant image is a combination between the original image and a filter that depends on the local content of the original image. It means that regions containing the same phases (low grey-scale gradient) are well smoothed while the edges of different phases (high grey-scale gradient) are retained.

The non-local means (NLM) filter has been developed to capture edges as well as finer details in inner phases (Buades et al., 2005; Buades et al., 2008). This filter evolves based on the Yaroslavsky filter (Yaroslavsky, 2012), which averages similar pixels using a low grey-scale gradient according to the intensity distance of the image. The difference between NLM and other methods is that the similarity between pixels is more robust through using regional comparison, which is not limited to be local. Pixels which are far from the current pixel are not impacted (Manjón et al., 2008).

Comparing all these methods, the non-local means filter not only smooths the image but also preserves the edge between phases. For this reason, this method was used in this study to smooth the raw images and differential images.

3.2.4 Segmentation

After acquiring 3D grey-scale images, different phases should be identified correctly for further analysis and quantification of petrophysical properties. Various segmentation al-

gorithms have been developed, and the most common two used in this study are global thresholding and watershed segmentation.

Global thresholding is the simplest method for image segmentation which depends on the grey-scale value of each pixel. This segmentation method is effective with an image containing only two phases, even with a high signal-to-noise ratio. If there are more than two phases, the intermediate phase is easily misidentified, particularly at the phase boundaries.

To identify phase boundaries, the watershed segmentation method is more accurate (Shepard et al., 2004). The gradient map computed from the image is analogous to the height map of a catchment basin. The algorithm is performed using the following steps and shown in Figure 3.6: a new gradient image is generated by calculating the gradients between neighbouring voxels at first. Phase boundaries are related to the areas of high gradient and could be eliminated from the region seeding. Next the areas of low gradient are seeded by eye and the range of grey-scale values for each phase is ascertained. The watershed algorithm then grows the user defined seed basins successively up the intensity map into the high gradient regions. Voxel misidentification can be reduced in this way and phase boundaries can be identified.

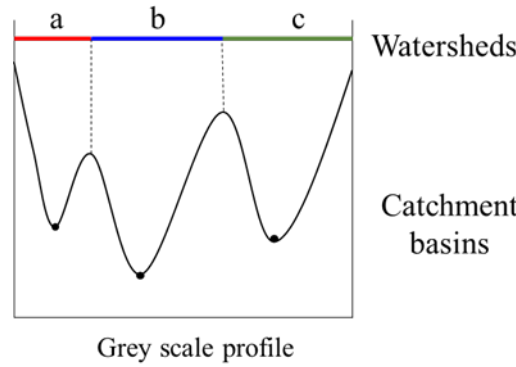


Figure 3.6: Generic watershed segmentation algorithm based on the grey-scale profile on one image (Sonka et al., 2014).

It is known that watershed segmentation method is suitable for two-phase segmentation with clear interfaces, for example, between solid phase and macro-pore phase, but it is not good for sub-resolution pore space with intermediate grey-scale values. Thus, in this study, global thresholding, which is based on the histogram of the grey-scale image, was applied in the segmentation for different phases in the Estailades carbonate sample, with the results of shown in Section 5.3.2 and Section 5.3.3, while watershed segmentation method was used

for segmenting phases with obvious interface, such as the identifying oil and water phases.

3.3 Principles of the differential imaging method

This method has been used for subvoxel analyses of datasets with changes that are not segmentable at the resolution of the image such as micro-porosity and intermittency. For example, the resolution of the micro-CT is not sufficient to identify explicitly pores that are smaller than a few microns across in this study. However, by analysing the change in grey-scale value in each successive image in a time series it is possible to discern changes in structure and fluid arrangement that would not otherwise be quantifiable. After the images are registered and resampled, the difference between the images is calculated.

The use of differential imaging between the brine saturated scan and the multiphase scan identifies clearly changes in fluid occupancy. The difference image between a scan where the rock is fully saturated with a high-contrast brine scan and the multiphase scan is defined as:

$$CT(differential) = CT(brine\ scan) - CT(multiphase\ scan) + CT0 \quad (3.5)$$

where CT is the CT number of each voxel, and $CT0$ is a constant value to avoid negative grey-scale values.

In this thesis, the differential imaging method is used in a novel application to: 1) identify intermittency in macro-porous space; and 2) calculate the oil recovery from micro-porous space, whose size is lower than the voxel size of micro-CT image.

3.4 Measurement error

To quantify potential measurement error for the image analysis process we conducted a sensitivity analysis of porosity on two of our sample rocks. We addressed the impact of gradient threshold in watershed segmentation on porosity identification and then we assessed how saturation changes as a result of these differences.

From this analysis we concluded that the measurement error associated with segmentation

using watershed is largest with more complex samples. However, the error is not so large as to deter the use of the watershed module, which is still the most widely used method for constructing pore-scale images. Segmentation is by nature subjective and care must always be taken in choosing gradient magnitudes that accurately reflect the rock properties of interest in the final image. When the results are presented later in the thesis, the degree of uncertainty introduced by the segmentation will be quantified.

3.5 Summary

In this chapter, the image acquisition and processing methods used in this work have been explained. In the next chapter, we will describe how the flow experiments with micro-CT imaging were performed to capture spatially resolved images during simultaneous two-phase flow at steady state.

Chapter 4

Experimental methodology

This chapter describes the experimental methodologies including the two-phase flow rig design and the X-ray imaging used to capture oil and water flow behaviours.

4.1 Lab-based micro-CT experiments for multiphase flow at steady state

Section 4.1.1 describes the design of the flow rig that is used in the micro-CT scanner. Section 4.1.2 introduces the procedures used to take scans at different fractional flows and to measure relative permeability.

4.1.1 Two-phase flow rig design

This section introduces the two basic flow loops used in the micro-CT: the co-injection flow rig and the dual-injection flow rig.

4.1.1.1 Co-injection flow rig

This flow loop is shown in Figure 4.1. It was constructed specially for the experiments on Bentheimer sandstone presented in Section 5.2. This apparatus consisted of a Zeiss Versa XRM-500 micro-CT, two ISCO syringe pumps for injecting oil and brine, one ISCO pump for keeping confining pressure, one ISCO pump for maintaining back pressure to the whole system, a lab alliance barrel wash pump, a precise pressure difference transducer, with several valves and tubes to control the fluid flow. The experiments were conducted in a Hassler type flow cell made of carbon fibre that is X-ray transparent. The rock sample was placed into a fluoropolymer elastomer (Viton) sleeve. Two metal fittings were attached to both sides to connect the core to the pore-fluid flow lines. Figure 4.2 illustrates the assembled core holder. Oil and brine were injected simultaneously into the core sample through a T-junction. Figure 4.3 shows how the flow loop is arranged in the micro-CT scanner. The core holder was located in the clamp on the stage, which rotated with the stage during the scan. Flexible peek tubing (Kinesis, St. Neots, UK, Part Number: 1533XL) was used in the flow loop to make sure the lines were easy to rotate with the stage and provide little lateral load on the core holder.

4.1.1.2 Dual-injection flow rig

The flow loop is shown in Figure 4.4. This was constructed specially for the experiments on Estailades limestone described in Section 5.3. The difference between these two flow rigs is the injection port. In this dual-injection design, brine and oil reach the sample simultaneously. The details are shown in Figure 4.5. The micro-CT scanner used in this study was a Zeiss Versa XRM-510 micro-CT with a flat panel, as shown in Figure 4.6. All the other facilities included in the flow loop were the same as the one shown in section 4.1.1.1.

4.1.2 Experimental procedures

The experiments were conducted using the following procedures:

1. Leak testing the flow loop. Before connecting the core holder, the flow loop should be

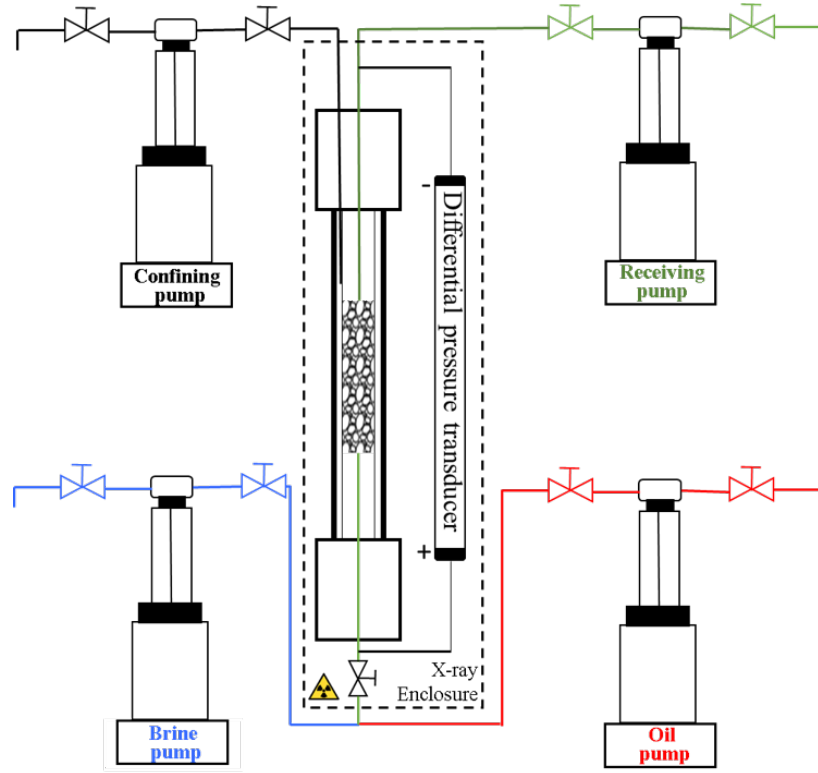


Figure 4.1: Co-injection experimental apparatus used in the Versa XRM-500. The pumps used to control the flow and the siting of the flow cell within the micro-CT enclosure are shown.

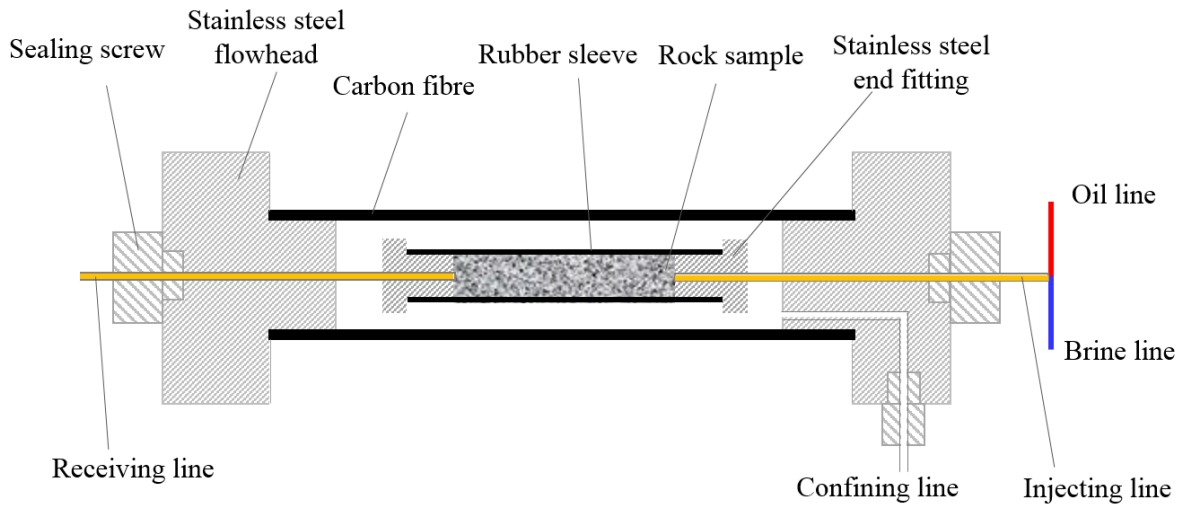


Figure 4.2: Detail of the core-assembly of the co-injection core holder made of carbon fibre, which is transparent to X-rays. Oil and brine were injected simultaneously through a T-junction.

tested to ensure that there was no leakage during the experiment to damage the micro-CT and other facilities. All parts of the apparatus were connected well and water was

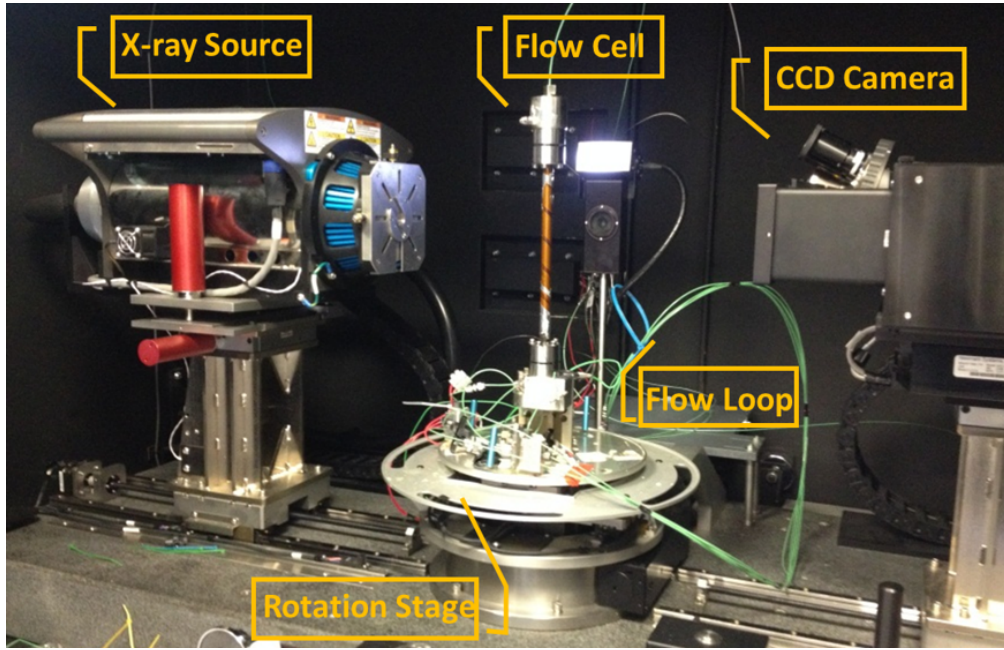


Figure 4.3: The laboratory-based Zeiss Versa XRM-500 micro-CT scanner consists of three main components: an X-ray source, a rotation stage which mounts and controls the sample position and an X-ray detector.

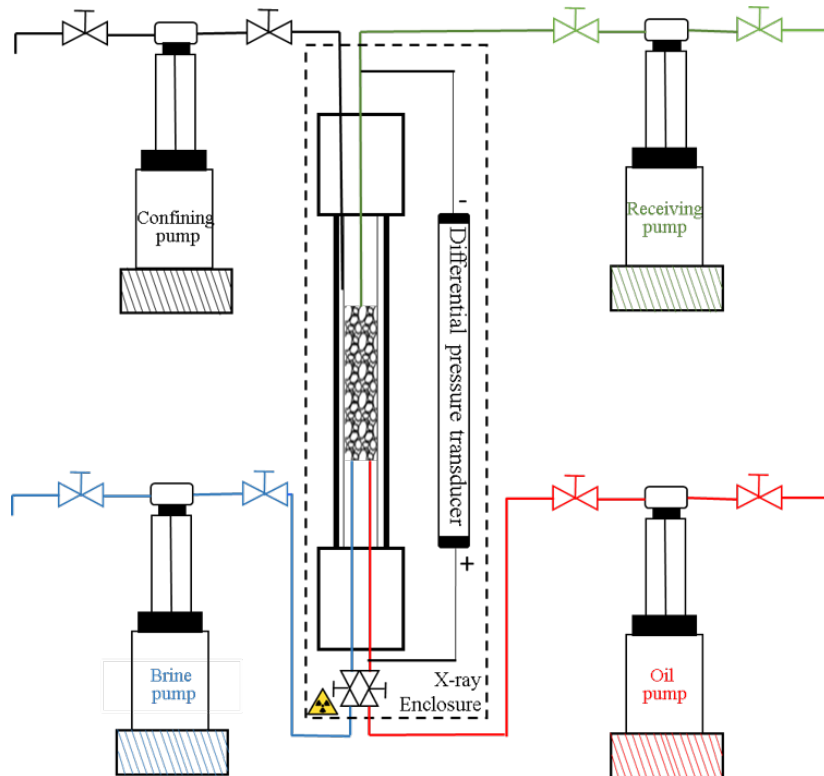


Figure 4.4: Dual-injection experimental apparatus used in the Versa XRM-510. The pumps used to control the flow and the siting of the flow cell within the micro-CT enclosure are shown.

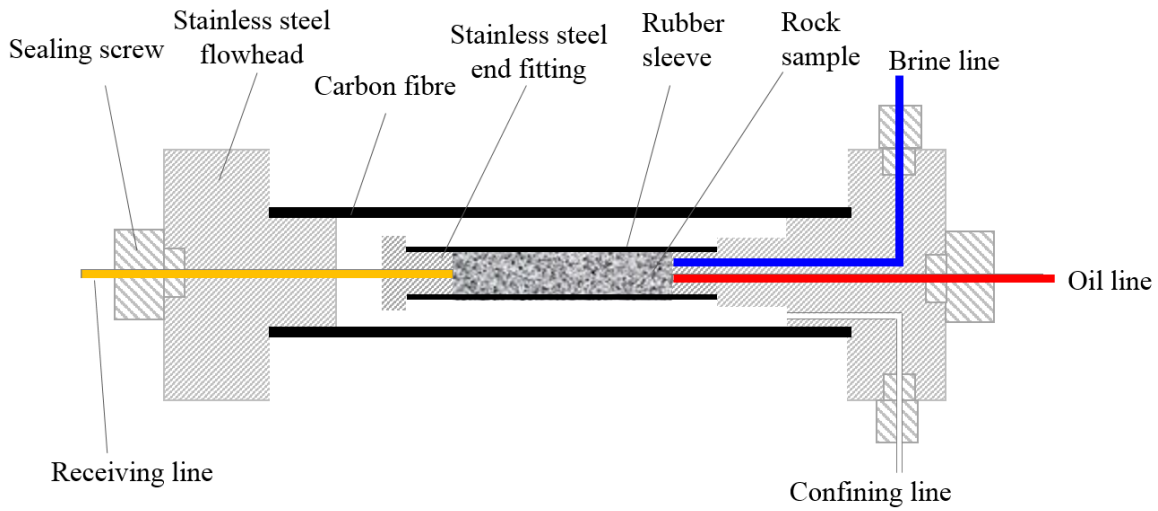


Figure 4.5: Detail of the core-assembly of the dual-injection core holder. Two separate ports guarantee that oil and brine reach the rock sample at the same time.

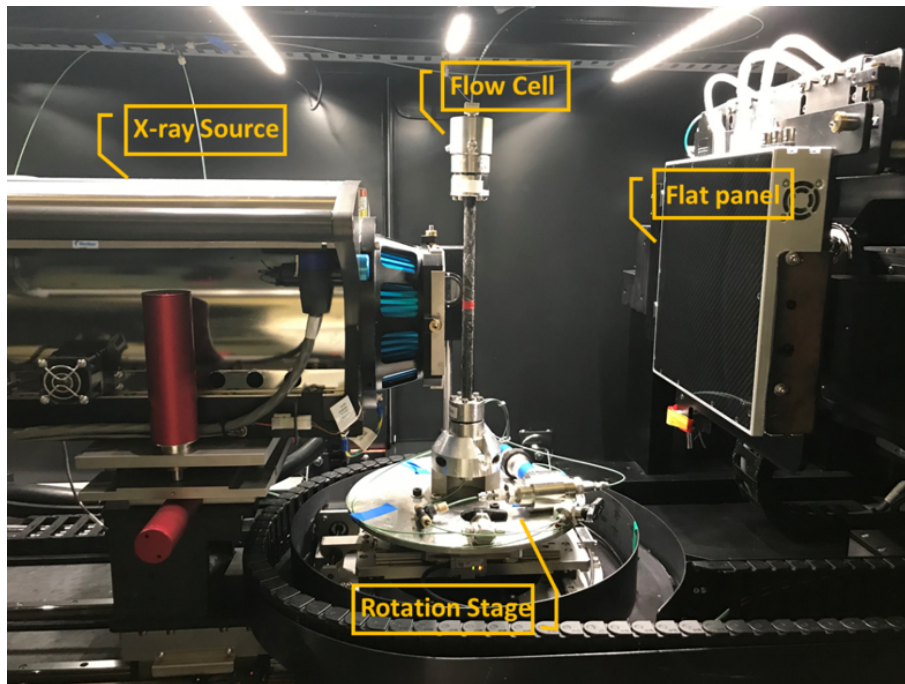


Figure 4.6: The laboratory-based Zeiss Versa XRM-510 micro-CT scanner with the flat panel inside.

injected into the whole system. The stage was moved out from the micro-CT and all the connections were kept closed. Then the pressure increased from 0 to 10,000 kPa step by step. Volumes and pressure of the injection pumps, receiving and confining pumps were recorded for 30 minutes to check for leaks.

2. Load the sample into the core holder. The core was loaded into the cell in preparation

for the flow experiment. The diameter of Bentheimer sandstone was 5 mm because the size of the end fitting was 5 mm. The sleeve was cut to size to create a tight seal to avoid bypass. The diameter of the Estailades carbonate was 6 mm because the size of the dual-injection end piece was 6 mm. There must be no space between the core and end fitting to ensure the confining pressure does not over compress the sleeve and pinch off flow. Aluminium tape was wrapped the sleeve and the end fittings to keep the seal. Finally, the core holder was fully assembled with the end caps.

3. The assembled core holder was mounted on the stage as Figure 4.3 shows for Bentheimer sandstone and Figure 4.6 shows for Estailades carbonate. The flow lines were connected by PEEK connectors. A differential pressure transducer (PD-33X, Keller) was linked along the tubing. The “+” linked to the tubing at the inlet of the core holder and the “-” linked to the tubing of the outlet of core holder. The range of the differential pressure transducer was 300 kPa for Bentheimer sandstone, and the accuracy was 0.1% of the whole range. The range of the differential pressure transducer was 5 MPa for Estailades carbonate, and the accuracy was 0.05% of the whole range. Care must be taken to ensure the core holder was completely vertical to allow the micro-CT source to be positioned as close as possible to decrease the scan time. At this stage it was also prudent to test the stage rotation and ensure that all flow and electrical lines are free to rotate from - 180° to 180°.
4. PEEK tubing was used to link the confining pump and the confining port of the core holder. A confining pressure of 2 MPa was applied and maintained within the cell to compress the Viton sleeve around the core sample to avoid any fluid bypass.
5. A dry (air) scan of 1 hour was taken at room temperature. A dry scan of the sample was taken before the whole experiment, which was used to image the pore space. The sample was put in the Viton sleeve and assembled in the core holder made of carbon fibre. The core holder was fixed on the stage. The energy was chosen based on an ideal transmittance between 0.2 to 0.3. The exposure time was chosen based on an average value of around 6000 counts of the image, which ensures that the signal to noise ratio was high and the detector was not over saturated. The dry scan was taken with around 2000 projections to provide good phase contrast and edge sharpness.
6. The core was flooded with gaseous CO₂ for 30 minutes to remove air.

7. 30 wt% brine was injected. The brine solution was made from deionised water with a prescribed amount of potassium iodide (KI). KI was used as an ionic salt as it has a high atomic weight causing a high X-ray attenuation, allowing it to be used as a contrast agent. A solution of 30 wt% KI in deionised water was used as the wetting phase, which provided an effective contrast between brine, oil and rock phase (Lin et al., 2016). It was checked that there was no air in the tubing linking between the main flow line and transducers. Additionally, as high salinity brine was used in the whole experiment, the pump was easily damaged if salt crystals precipitated inside the barrel, resulting in scratching the interior of the pump and fluid leakage along the seals. Thus the brine injection pump should be constantly washed with a barrel wash at the same time, in which deionised water is constantly circulated along the outer edge of the barrel to keep any potential precipitates in solution.
8. Brine was injected from the brine pump at a high flow rate (1 mL/min) for more than 5 hours through the core sample to ensure it was fully saturated with brine with no CO₂ in the system.
9. After 100% brine saturation was achieved, a brine saturated scan for 1 hour was taken. Image acquisition followed the same procedure as Step 5.
10. The first step was primary drainage. Only oil was injected into the core to obtain the initial water saturation. For the low Ca experiment, (see step 16 for the high Ca experiment) the two pumps were 100DX High Pressure Syringe Pump (Teledyne Isco Syringe Pumps). The flow range was from 0.01 μ L/min to 50 mL/min, and the flow accuracy is $\pm 0.3\%$ of setpoint. The total low flow rate was 0.02 mL/min.
11. Wait until the flow reached steady state. It took about 20 hours to reach steady state at low Ca to see stable conditions, which were identified by observing a stable and constant pressure drop. The scanning time depends on several factors including voltage and exposure time and should be optimized. The longer the time for scanning, the better the image quality is. However, fluids flow faster than the micro-CT 3D scan, the longer time for scanning, less dynamic phenomena can be captured. Thus it is important to balance the scan time and image quality. After taking images at different projections, we compared the image quality and dynamic fluid behaviours. The exposure time was adjusted to reach at least 6000 counts over 7 s so that there was an adequate signal to noise ratio. Between 1400 and 1600 projections were used per tomography. To reduce

scan time further, auto referencing was disabled and the only reference used was the one taken separately before each scan. Additionally, sample drift correction was disabled as the time and spatial resolution of these scans is such that drift correction did not improve quality. To avoid the source colliding with the core holder, the reference axis was set to $-X$ and the sample too large for 'auto ref' box was ticked. These steps should be followed strictly for every tomography all the time as the default setting is not suitable for scanning with a large core holder inside the micro-CT and can indeed be dangerous if used improperly.

12. We chose to take approximately 1 hour for the scan for the Zeiss Versa XRM-500 micro-CT scanner and 40 minutes in the Zeiss Versa XRM-510 micro-CT scanner without stopping fluid injection.
13. Steps 10 and 12 were repeated at water fractional flows of 0.15, 0.3, 0.5, 0.7, 0.85 and 1 by changing the flow rates of the oil and brine pumps while keeping the total volumetric flow rate constant. The oil flow rate decreased from 0.02 mL/min to 0 mL/min and the brine flow rate increased from 0 mL/min to 0.02 mL/min. The experiments therefore represent an imbibition (waterflood) displacement. The average pressure difference at each fractional flow was recorded.
14. After the last scan finished, the whole system should be efficiently depressurized. All the injection pumps were stopped. The pressures of the confining pump and the receiving pump were decreased one by one and step by step to avoid breaking the sample. Once the pressure had declined to around 100 kPa, the confining and receiving pumps were opened to the atmosphere.
15. The core was cleaned by injecting more than 1000 mL deionised water at 1 mL/min for more than 5 hours and put in an oven at 80°C for 12 hours to dry it. A quick dry scan could help to ensure the sample was clean enough.
16. Steps 2 to 14 were repeated with a total flow rate of 0.5 mL/min ($Ca = 7.5 \times 10^{-6}$) for the Bentheimer sandstone. A total flow rate of 0.7 mL/min ($Ca = 7.3 \times 10^{-6}$) was applied on the Estailades carbonate. It took about 10 - 15 hours to reach steady state in this case. For this high Ca experiment, the two pumps were 1000D High Pressure Syringe Pump (Teledyne Isco Syringe Pumps). The flow range was from 0.1 mL/min to 408 mL/min, and the flow accuracy is $\pm 0.5\%$ of setpoint. The reason we chose a larger

volume pump was that the volume of the 100D pump was too small (102 mL) to flood constantly for 15 hours.

17. After collecting all the pressure drops, the system was depressurized. The sample was taken out and the pressure difference during flow in only the tubing between the inlet and outlet at each fractional flow at low and high Ca was measured by repeating the steps 10 to 13 and step 16. For low Ca , the pressure drop along the tubing was from 0.1 kPa to 0.9 kPa. For high Ca , it was from 2 kPa to 21 kPa. The pressure difference was the smallest when f_w is 0, and was the largest when f_w was between 0.5 and 0.75.
18. The pressure drops in the cores were calculated by subtracting the pressure drops along the tubing measured at step 17 from the total pressure drops measured at steps 10, 13 and 16.
19. After finishing the experiment, all the pumps were stopped and washed. The brine pump was cleaned by DI water three times. The oil and receiving pumps were cleaned by Acetone twice and DI water three times. All the tubing was taken out of the micro-CT. The core was taken out of the core holder and the sleeve disconnected from the interior end fittings.

4.2 Synchrotron tomography experiments

All imaging was performed at the Diamond Imaging Beamline I13 (Diamond Light Source, UK), as shown in Figure 4.7. The polychromatic light of the I13 beamline at Diamond Light Source synchrotron in Didcot, UK, was used to image the core. Figure 4.8 shows the apparatus used at the Diamond Light Source (DLS) Beamline I13. There are two main differences between scanning at Diamond light source beamline and scanning in the lab-based micro-CT: energy quantity and energy type. The total energy flux using the Diamond pink (polychromatic) beam is much greater than that of the micro-CT and thus a tomography that would take 30 - 100 minutes in the laboratory only takes approximately 1 minute at Diamond light source beamline. This allows for a much better time resolution and to some extent, a better quality image. The imaging apparatus at Diamond light source beamline consists of a polychromatic X-ray beam produced by an undulator. Thus, the flux is highly quantized over a narrow spectrum. The energy of the beam ranges from 8 KeV



Figure 4.7: Synchrotron facility at the Diamond Light Source (<https://www.diamond.ac.uk/Home/About.html>).

to 30 KeV. As the lower energy X-rays cause heating of the sample and do not add to image contrast, they were filtered out using a 0.2 mm pyrolytic carbon filter, 2.2 mm of aluminium and 0.1 mm of gold. The detector was composed of a 250- μm -thick cadmium tungstate (CdWO_4) scintillator with a $1.25\times$ objective lens and a pco.edge scmos (scientific complementary metal-oxide-semiconductor) image sensor camera.

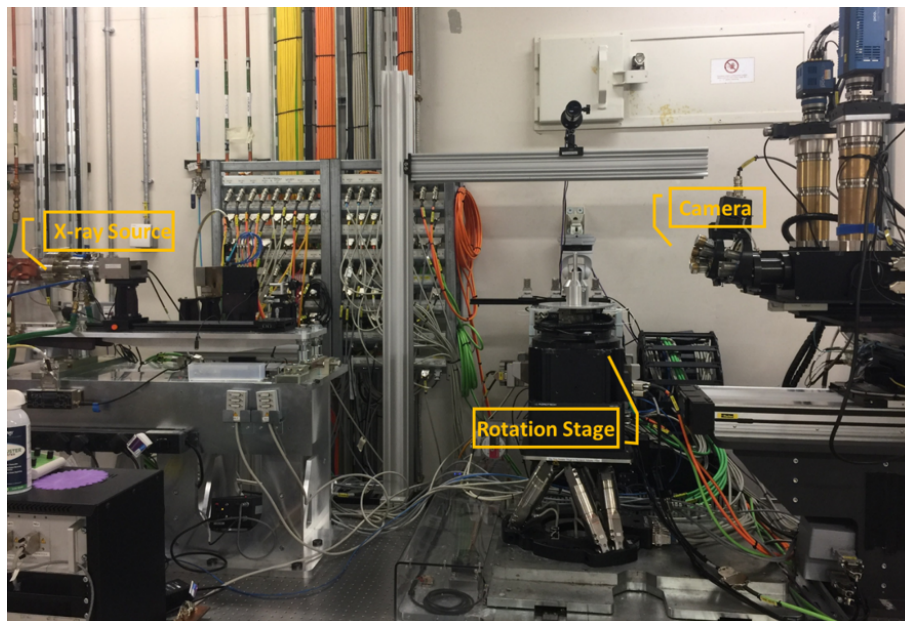


Figure 4.8: Apparatus used at the Diamond Light Source (DLS) Beamline I13.

4.2.1 Experimental protocol

The experimental apparatus used at the Diamond Light Source (Figure 4.9) was nearly the same as the one used inside the micro-CT.

The main difference was that the special core holder which was designed to sit on the rotation stage was much shorter than the one used inside the micro-CT. This is because the stage at the Diamond synchrotron rotates much faster than the one in the lab-based micro-CT and the core holder made of carbon fibre is relatively soft and flexible. Thus, a shorter core holder was which used less prone to bend and flex. This minimized possible motion effects, such as sample movement which would distort the images.

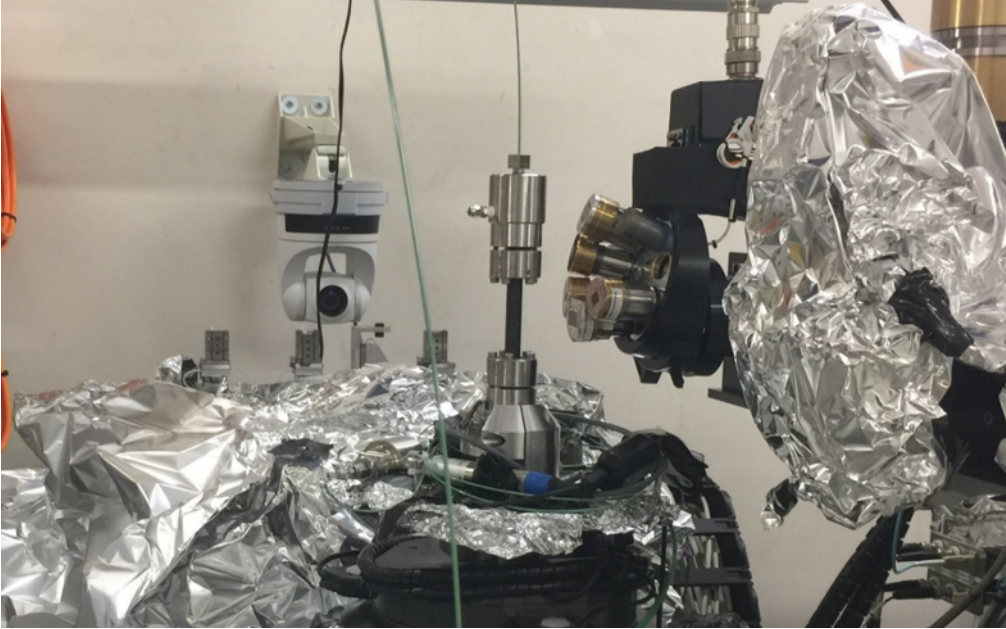


Figure 4.9: The core assembly mounted on the rotation stage. The body size of the core holder was much shorter than the one used in lab-based micro-CT. Flow and electrical lines were free to rotate. Aluminium foil was used to cover the stage and lens that were not used, to prevent any potential damage caused by leakage.

By taking advantage of the quick scan property of synchrotron, we simultaneously flowed both the wetting fluid (brine) and the non-wetting (oil) at independently controlled volumetric flow rates to investigate multiphase flow. The dynamic flow displacement experiments were performed using the procedures described below:

1. Choose the optimum setting parameters for scanning. A sample was located on the rotation stage. The distance between the sample and X-ray source, the distance between

the sample and camera, projection numbers and exposure time were determined to balance the image quality and scan time. In this study, images were acquired using 1,000 projections with an exposure time of 0.04s. The total acquisition time for each image was 1 min: 40 s to take the projections and around 20 s to transfer the data from camera to computer and to rotate the sample back to the starting position. The number of projections was chosen to optimize the resolution of the final images without significantly increasing the acquisition time.

2. Contrast test scans were taken before the experiment. Bentheimer sandstone powders were divided into 7 parts equally and put into 7 small glass bottles. 10 wt%, 15 wt%, 20 wt%, 25 wt%, 30 wt%, 40 wt% , and 50 wt% KI brine were prepared and poured into the bottles respectively. Then decane was put inside each bottle. These materials were well mixed. Quick scans were taken for these salinities. By comparing the contrast of the oil, brine and grain phases, 15 wt% KI brine was chosen as the optimum brine concentration.
3. Leakage test of the flow loop. The procedures were as the same as for the lab-based micro-CT scanner, section 4.1.2.
4. A dry scan was taken with 2 MPa confining pressure, which compressed the Viton sleeve outside the core to avoid fluid bypass.
5. Air was displaced by CO₂ by injecting CO₂ into the core sample for half an hour.
6. 15 wt% brine was injected into the sample to make sure the sample was 100% saturated with brine. A differential pressure transducer (PD-33X, Keller) was installed along the tubing. The “+” linked to the tubing at the inlet of the core holder and the “-” linked to the tubing of the outlet of core holder. The tubing linking the main flow line and the transducer remained full of brine during the experiment. It was assumed that the transducer measured the pressure difference in the oil phase. The range of the differential pressure transducer was 500 kPa, and the accuracy was 0.05% for the whole range.
7. The brine-saturated sample was scanned. A back pressure regulator was set to 2,000 kPa for the whole system.
8. Oil was injected at 2 mL/min for 30 minutes to reach a low initial water saturation.
9. Then water and oil were injected when f_w were 0.15 and 0.3 by keeping the total volumetric flow rate fixed at 0.02 mL/min. Each fractional flow was injecting for one and

half hours.

10. Water and oil were injected at equal flow rate of 0.01 mL/min respectively. At the same time, the pressure drop across the whole sample was recorded during the experiment. We waited a further two hours after the pressure stabilized. 3D images were taken all the time.
11. The total flow rate was increased to 0.04 mL/min, 0.08 mL/min, 0.4 mL/min, 0.8 mL/min, 1.2 mL/min, 2.5 mL/min and 4 mL/min step by step while fractional flow was kept at 0.5. For each flow rate, it took two hours until steady state was reached. Successive scans were taken from the start without stopping. The images were time-averaged over one minute.
12. The system was depressurized when the experiment finished. The sample was taken out and the pressure difference during flow in only the tubing between the inlet and outlet at each flow rates was measured by repeating the same steps from step 7 to step 11.
13. The pressure differences along the cores were calculated by subtracting the pressure drops along the tubing measured at step 12 from the total pressure drops measured from steps 10 to 11.

4.3 Measurement error

We consider the errors caused by length and diameter measurement, the accuracy caused by pumps, the accuracy of the precise differential pressure transducers. The details of these are listed in Table 4.1. The range and accuracy listed in the table are obtained from manual book of facilities. Errors are added to pressure measurement.

Table 4.1: Details of the accuracy of the facilities

Equipment	Brand and type	Accuracy	Range
Pumps 1000D	Teledyne Isco Syringe Pump	$\pm 0.5\%$	0.001 - 408 mL/min
Pumps 100D	Teledyne Isco Syringe Pump	$\pm 0.3\%$	0.01 μ L/min - 50 mL/min
Pressure transducer 3 bar	PD-33X, Keller	0.1%	0 - 300 kPa
Pressure transducer 5 bar	PD-33X, Keller	0.05%	0 - 500 kPa
Pressure transducer 50 bar	PD-33X, Keller	0.05%	0 - 5 MPa

4.4 Summary

The principles of micro-CT imaging and details of the novel experimental procedures were demonstrated in this chapter. The following three items are novel, developed and applied here to study multiphase fluid flow at the pore scale using X-ray imaging:

- 1). This is the first time that a differential pressure transducer was used in pore-scale experiments to measure the pressure drop across a mm-scale sample. This was used to interpret pore-scale dynamics and measure relative permeabilities.
- 2). A dual-injection end piece was designed specifically for this experiment to avoid any slug-flow in the sample.
- 3). The images were taken without stopping the flow. Compared with other experiments at static conditions, images taken from these dynamic experiments can show how immiscible fluids flow at steady state.

In the next chapter, the experimental results for steady-state flow in both Bentheimer sandstone and Estailades carbonate will be presented, using a lab-based micro-CT scanner.

Chapter 5

Pore-scale relative permeability experiments on Bentheimer sandstone and Estailades carbonate and the role of intermittency

In this chapter, pore-scale relative permeability experiments on Bentheimer sandstone and Estailades carbonate, and the role of intermittency are introduced.

5.1 Rock properties

To investigate the effect of rock type and pore-scale heterogeneity, two rock samples are examined in this thesis: Bentheimer sandstone and Estailades carbonate. We chose these samples because they are readily available, diverse in pore structure (Tanino and Blunt, 2012; Bijeljic et al., 2013a,b), relatively chemically pure and there is some previously-published literature containing experimental results for comparison.

5.1.1 Bentheimer sandstone

Bentheimer sandstone comes from a shallow marine formation deposited during the Lower Cretaceous, an example of which can be found in the Netherlands and Germany (Peksa et al., 2015). Since Bentheimer has good rock properties such as lateral continuity and is homogeneous at the metre-scale, it is an ideal sedimentary rock for reservoir studies. It is a type of angular quartz arenite with well-sorted medium grains. This makes it suitable for standard laboratory experiments and associated comparison with theory (Klein and Reuschlé, 2003). Based on the study performed at the Natural History Museum (London, UK), the samples we studied consisted of approximately 95% quartz, 4% feldspar and 1% fine clay (Andrew, 2014), shown in Figure 5.1.

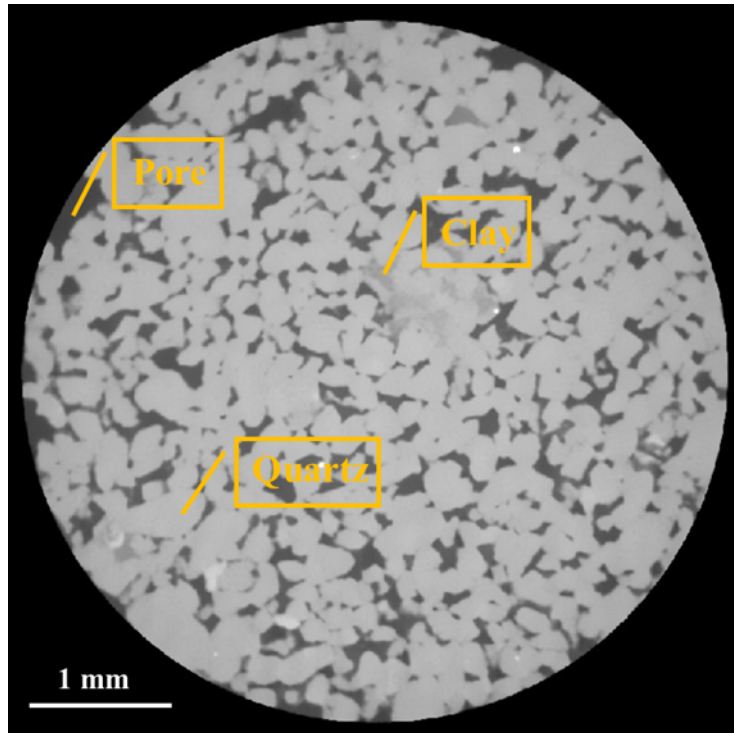


Figure 5.1: Two-dimensional cross-section of a 3D micro-CT image of the Bentheimer sample with a voxel size of $6\text{ }\mu\text{m}$. The majority of the grains are composed of quartz with only a small amount of clay.

The mercury injection capillary pressure was measured by Weatherford labs, East Grinstead. The results are shown in Figure 5.2 and the throat size distribution inferred from the capillary pressure is shown in Figure 5.3 (Blunt, 2017). Most throats – or restrictions in the pore space – have a radius greater than $10\text{ }\mu\text{m}$, which is well above the resolution of our images. This shows that for this sample we can explicitly resolve most of the pore

space.

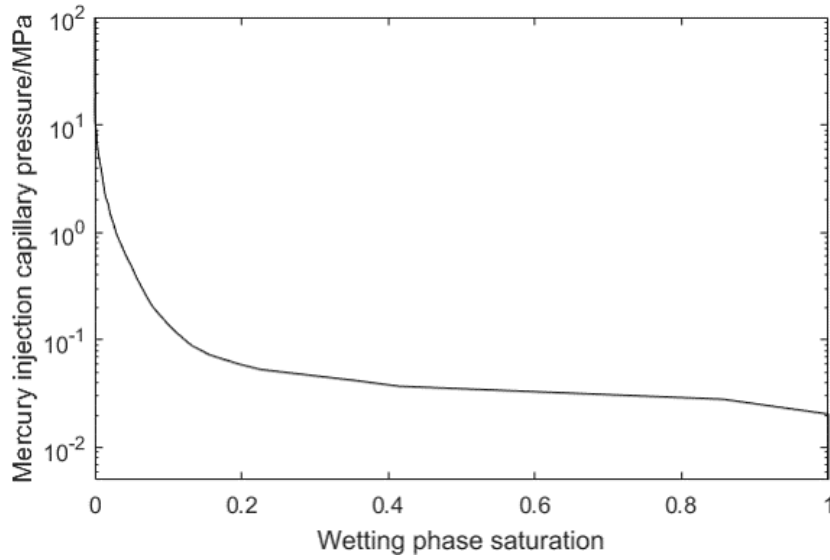


Figure 5.2: Mercury injection capillary pressure for the Bentheimer sandstone sample.

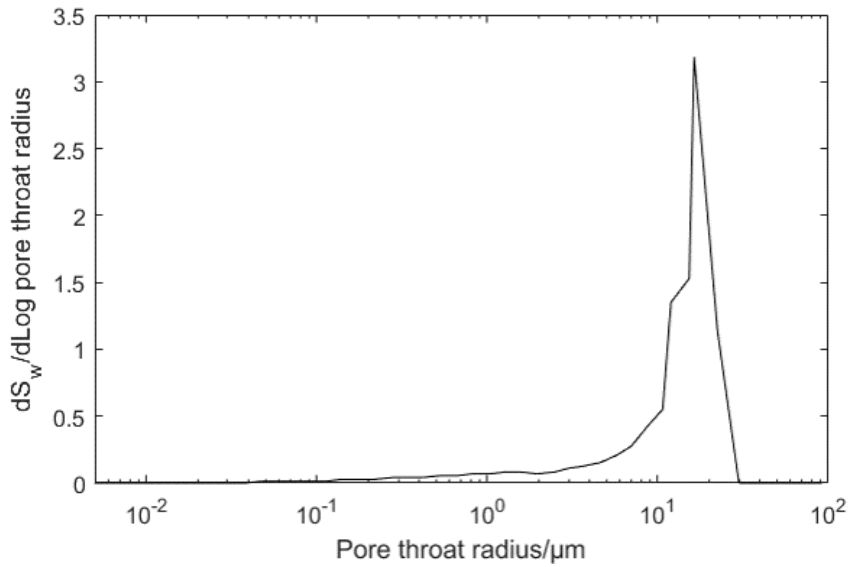


Figure 5.3: Throat size distribution for the Bentheimer sandstone sample inferred from the measured capillary pressure.

5.1.2 Estailades carbonate

Estailades limestone is a beautiful white limestone rock, quarried near the town of Oppède, France (<https://www.sculptures-bidal.com/en/article/10/our-stones/technical-information->

sheet-for-the-estailades-stone), and was deposited 22 million years ago. It is a medium to coarse grained moderately sorted bioclastic grainstone, which is almost universal calcitic with a minor quartz component. The allochem population is principally bioclasts (70 %) with a significant population of angular to subangular calcitic grains. The bioclast population consists of bivalve molluscs, bryozoa and coral fragments, as shown in Figure 5.4.

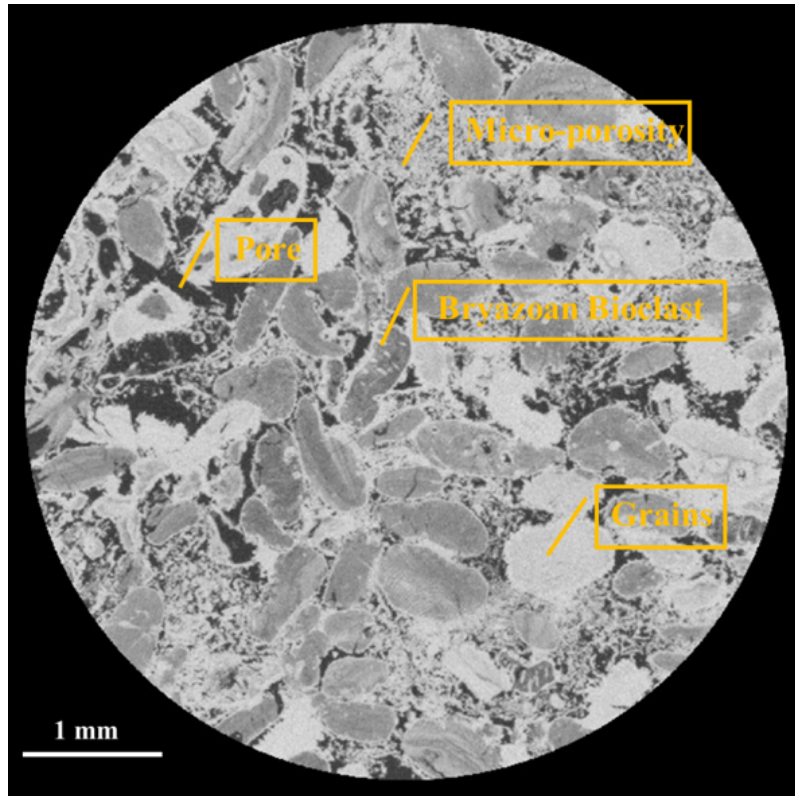


Figure 5.4: Two-dimensional cross-section of a 3D micro-CT image of the Estailades sample with a voxel size of $3.6\mu\text{m}$.

The mercury injection capillary pressure shows two distinct populations of throats, see Figure 5.5. Figure 5.6 shows the inferred throat size distribution: approximately half the pore space has radii in excess of $1\mu\text{m}$, which can be imaged, but there is a significant fraction of the porosity that is well below our image resolution. Fluid occupancy in these pores can only be assessed using differential imaging.

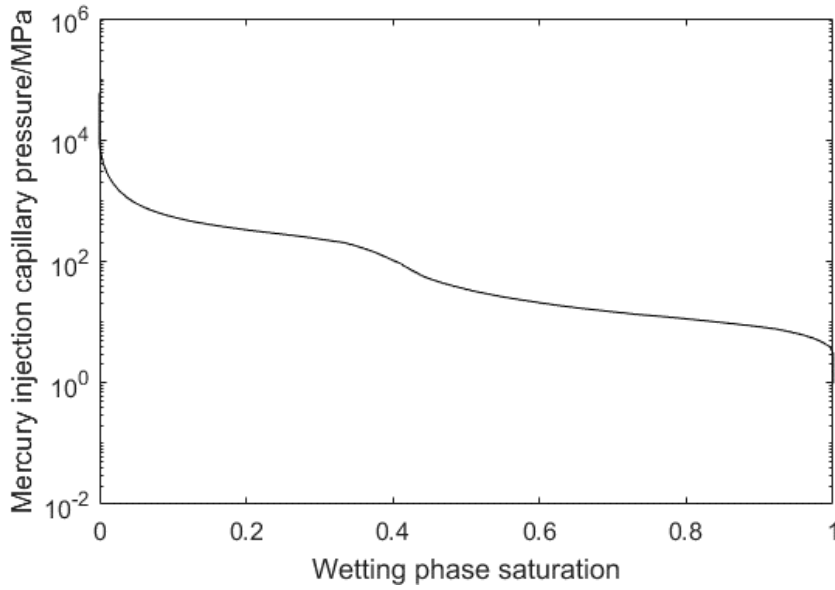


Figure 5.5: Measured mercury injection capillary pressure for the Estailades limestone sample.

5.2 Pore scale two-phase flow steady-state experiment in Bentheimer sandstone

This work has been published in Gao et al. (2017).

5.2.1 Introduction

Fundamentally, multiphase flow is controlled by the interplay of viscous and capillary forces whose balance depends on the geometry and topology of the porous structure, wettability, gravity, and flow regime (Blunt, 2017). Different flow regimes on the pore scale are characterized by the ratio of viscous to capillary forces conventionally described by the capillary number, $Ca = \mu q / \sigma$, where in this work μ is the mean viscosity of the flowing phases, q is the total Darcy velocity, and σ is the interfacial tension. However, this does not represent a true ratio of forces during a displacement (Hilfer and Øren, 1996; Armstrong et al., 2014): later in the section we will discuss the application of macroscopic measures of the balance of capillary and viscous forces.

In this section, we study the behaviour of oil and water phases over a range of fractional flows and combined laboratory imaging with sensitive pressure measurements which allowed

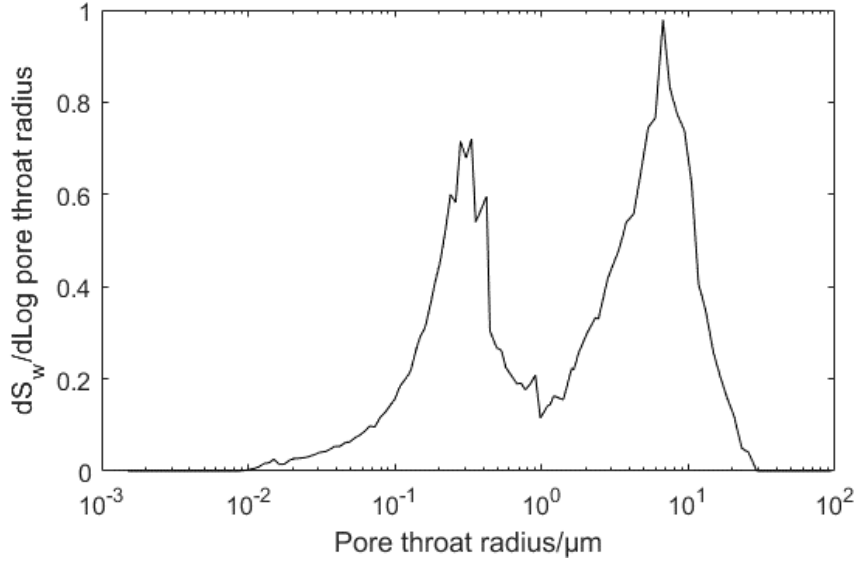


Figure 5.6: Throat size distribution for the Estailades limestone sample, showing two distinct populations.

the simultaneous estimation of relative permeability at the mm-scale. It is not possible to identify rapid changes in pore occupancy from comparing images, since these were taken an hour or more apart. Instead we devised a differential X-ray imaging method that can identify intermittent pore occupancy during simultaneous injection of wetting and non-wetting phases in the pore space of Bentheimer sandstone. We quantified the flow behaviour at two capillary numbers $Ca = 3.0 \times 10^{-7}$ and $Ca = 7.5 \times 10^{-6}$ representing two flow regimes over a range of fractional flows (0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1). We focus on two particular aspects: (i) to discriminate and quantify the intermittency and connectivity, and (ii) to measure relative permeability and compare our results with experiments on cm-scale samples.

The samples were drilled into cylindrical cores 4.99 ± 0.01 mm in diameter. Two samples were assembled in series, the bottom one was used as “diffuser” to mix the oil and water together, which helps ensure that two fluids flow into the second core simultaneously. The length of the two samples was 29.1 ± 0.01 and 30.9 ± 0.01 mm. The total length was therefore about 60 mm. The absolute permeability measured at the same experimental conditions was $2.045(\pm 0.056) \times 10^{-12}$ m². The porosity measured based on our image was 21.95 ± 0.52 %. A solution of 30 wt% KI in deionised water was used as the wetting phase, which provided the optimum contrast between brine, oil and rock phases (Lin et al., 2016). Decane was used as the non-wetting phase.

The interfacial tension (IFT) between brine and decane was measured to be 47.05 ± 1.56 mN/m at ambient conditions using a Ramé-Hart apparatus (590 F4 series) based on the pendant drop method (Andreas et al., 1938; Stauffer, 1965). The density of brine was measured to be 1263 ± 2 kg/m³ at ambient conditions by measuring the mass of a 1 mL drop of liquid. The accuracy of the mass balance is 0.001 g, and that of the volume pipette is 0.001mL. The density of decane is 730 kg/m³ (n-Decane, Acros Organics).

The flow regime depends on the fractional flow and the capillary number. In this work, the low total flow rate was 0.02 mL/min and the high total flow rate was 0.5 mL/min. The viscosity of brine is 0.822 ± 0.013 mPa·s, and that of decane is 0.838 mPa·s (provided by PubChem, open chemistry database). The capillary numbers, Equation (2.6), used in this work were calculated from the total flow rates of brine and decane, and the average viscosity of both fluids, because of their similar viscosity. The fractional flow is given by the injection rate ratio of each fluid to the total fluid rate: the capillary numbers for the low and high flow rate experiments were 3.0×10^{-7} and 7.5×10^{-6} , respectively. For each experiment we kept the total flow rate fixed as we varied the fractional flow.

We devised a differential imaging method to first examine the impact of sub-resolution pores in co-injection (Section 5.2.4) and then to quantify the intermittent flow during a co-injection experiment (Sections 5.2.5.1 and 5.2.5.2). In addition, the grey-scale distribution of the raw multiphase images was used to calculate the oil saturation (Section 5.2.5.3). Finally, the relative permeabilities were calculated based on the pressure drop measured between the inlet and outlet (Section 5.2.6).

5.2.2 Grey-scale images

The experimental procedures were followed using the steps described in Section 4.1. The rock and fluids were imaged using a Zeiss XRM-500 X-ray microscope at a voxel size of 6 μ m. The images used for analysis were cropped cylindrical sections to avoid artefacts at the boundaries of the core. The image size was $710 \times 710 \times 800$ voxels, or 4.26 mm diameter and length 4.8 mm, which represents a bulk volume of 68 mm³, or approximately three fifths of the total size of the imaged sample. After reconstruction, all the images were registered to the dry scan image to have the same orientation using normalized mutual information, and resampled using the Lanczos algorithm (Burger and Burge, 2016), as introduced in Section

3.2.2.

Figure 5.7 and Figure 5.8 show two-dimensional cross-sections of three-dimensional micro-CT images at low and high Ca respectively over a range of fractional flows for Bentheimer sandstone. Grey is the grain phase and intermittent oil, black is the oil phase and white is the brine phase.

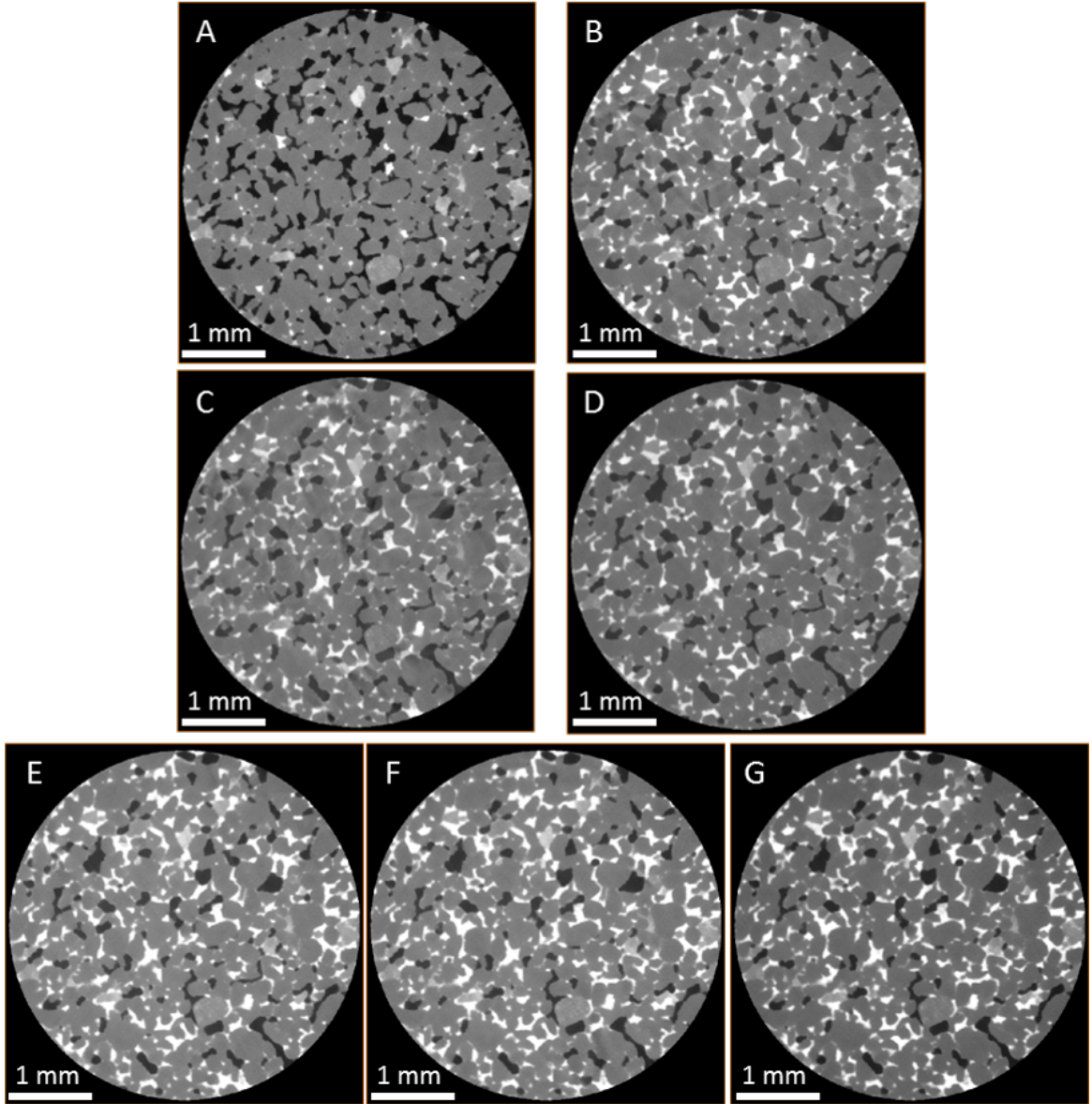


Figure 5.7: Two-dimensional cross-sections from three-dimensional micro-CT images at different fractional flows when Ca is 3.0×10^{-7} . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Grey is grain, black is oil and white is brine.

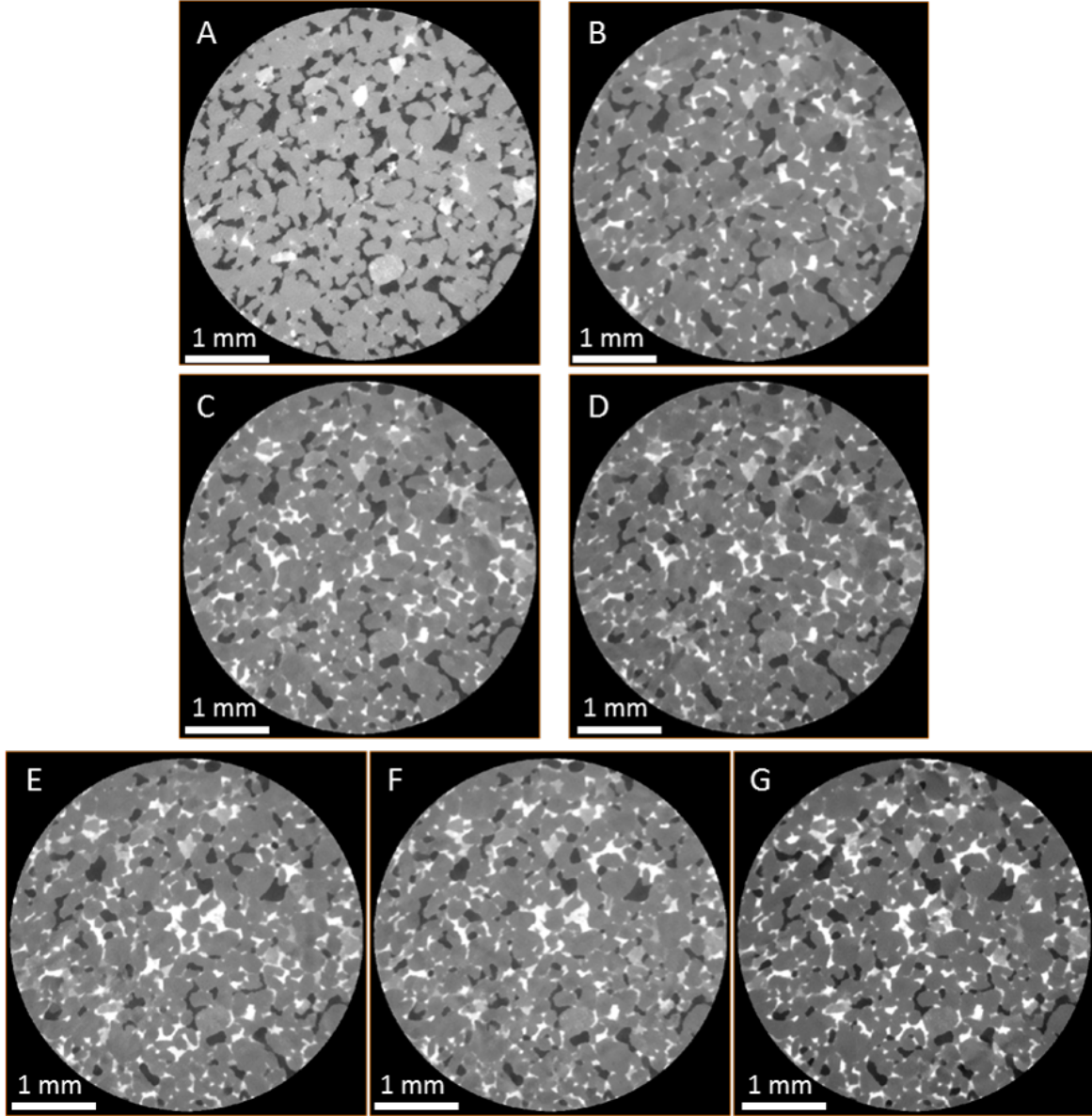


Figure 5.8: Two-dimensional cross-sections from three-dimensional micro-CT images at different fractional flows when Ca is 7.5×10^{-6} . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Grey includes grain and intermittent oil, black is stable oil and white is brine.

5.2.3 Differential images and dynamic connectivity at different Ca

Even though all the images were taken at the same scan conditions, the grey-scale value of each voxel was different because of varying fluid proportions and corresponding X-ray attenuation. Image normalization is a method to match the frequency distribution of the grey-scale histogram of the multiphase image with that of the brine image, which helps

to distinguish the different phases. It can avoid image artefacts and aid more accurate segmentation.

Using 30 wt% KI doped brine helps to maximize the phase contrast between the oil and water phases. The main aim of the differential imaging method is to enhance the phase contrast. 30 wt% KI has been used previously to visualize and quantify the sub-resolution porosity in carbonates (Lin et al., 2016). In this study, to quantify fluid intermittency, the brine phase is also doped with 30 wt% KI. In principle, either phase can be doped in order to enhance the phase contrast, e.g. the oil phase can be doped with iododecane. The reason for doping the wetting phase is because it should fully saturate the core initially and we can use this to check whether there is no air present in the brine phase at the start of the experiment. The image contrast between pure deionised water and air under X-ray is not good, especially in small pores. The use of differential imaging between the brine saturated scan and the multiphase scan identifies clearly changes in fluid occupancy.

The differential imaging method described in Section 3.3 was used to evaluate the saturation in sub-resolution pores, although for Bentheimer these only represent a small fraction of the pore space. Figure 5.9(A) shows the dry, brine and multiphase image when f_w is 0.5 at low and high Ca . Figure 5.9(B) shows the differential image at different conditions for Bentheimer.

At the lower capillary number, there is only black and white in the differential image, thus the non-wetting phase is transported in distinct connected pathways which do not appear to change during the hour-long scan time. However, at high capillary number, intermediate grey values are seen in the differential image (indicated by the red boxes in Figure 5.9(B2)), which suggests that the non-wetting phase is only present in these locations for some of the time during the scan at steady state – this suggests that there is intermittent behaviour because locally the pore-scale configuration of fluid rearranges.

5.2.4 The impact of sub-resolution pores during co-injection

The differential imaging method was first used to evaluate the saturation in sub-resolution pores. Based on the dry scan and fully brine-saturated image shown in Figure 5.9(A1) and (A2), some sub-resolution pores exist in Bentheimer. As Figure 5.9(B) shows, the grey re-

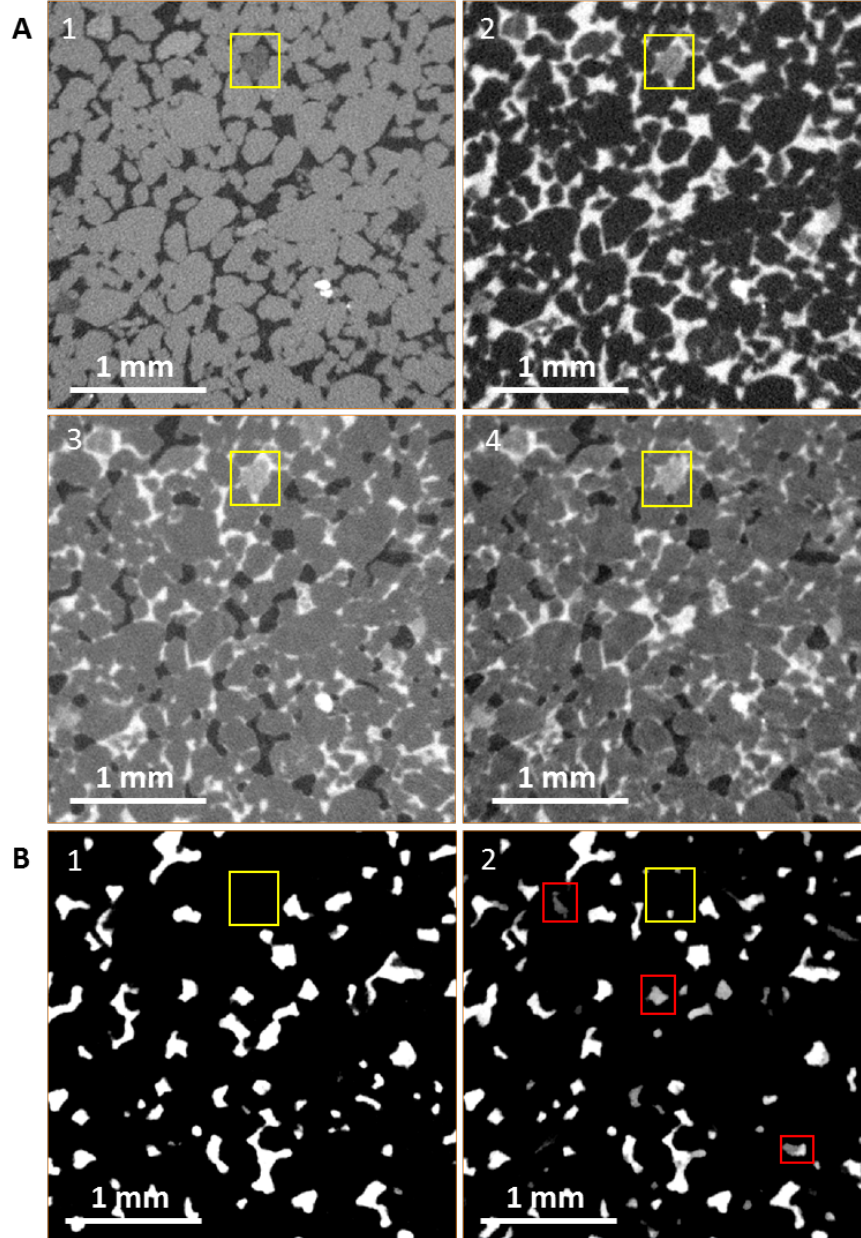


Figure 5.9: (A) Two-dimensional cross-sections of three-dimensional micro-CT images. (1) An example of a dry scan. (2) Sample saturated with brine. (3) Image after co-injection of oil and brine at low Ca when f_w was 0.5. (4) Image after co-injection of oil and brine at high Ca when f_w was 0.5. The yellow boxes indicate regions containing micro-pores. (B) The difference in grey-scale values between the KI saturated scans and the multiphase scan at low Ca (1) and high Ca (2) at steady state. The two-dimensional slices show that three phases including solid grain phase and unchanged brine (dark), redistributed oil (intermediate grey) and oil phase (bright) can be distinguished. The yellow boxes, see Figure 5.9(A), show that the sub-resolution pores are only occupied by brine. The red boxes show the presence of grey in the differential image indicating intermittent pore occupancy.

gions, indicated by the yellow boxes, are dark in the differential image, Figure 5.9(A), which confirms that oil does not enter these regions. The capillary entry pressure is insufficient

for the oil phase to displace brine from these small pores.

5.2.5 Quantification of phase saturation and occupancy

5.2.5.1 Image processing and segmentation-based differential imaging

The differential images were pre-processed by filtering with a non-local means edge preserving filter to reduce the image noise (Buades et al., 2005; Buades et al., 2008). This is a linear filter where the grey scale of a certain voxel is replaced by the average of the grey scales of other voxels. In contrast to the standard mean filter, each voxel within the search area is assigned a weight proportional to an index measuring the similarity of the weighted voxel to the voxel being replaced by the filter. Each voxel is essentially averaged with other voxels similar to it, including interface voxels, which means that edges are preserved. Then the filtered differential images were processed with global thresholding to identify brine, oil, and redistributed (intermittent) phases, as described below - for more details see Lin et al. (2016).

Segmentation of the intermittency follows the procedure shown in Figure 5.10. The pore space was segmented based on the differential image between the dry scan and the brine scan, shown in Figure 5.10(E). Pore thought to shows intermittent and oil phases, Figure 5.10(F), were segmented based on Figure 5.10(D). Stable oil, Figure 5.10(G), was identified based on the differential image. The intermittency was segmented by obtaining the difference between Figure 5.10(F) and Figure 5.10(G).

5.2.5.2 Quantification of the intermittency saturation

At the lower Ca , the oil saturation was obtained from the differential images directly as no intermittent flow was identified. The water saturation was calculated based on the total porosity of the whole sample and the fraction of the pore space occupied by oil. The total porosity, ϕ , was $21.95 \pm 0.52\%$, calculated based on the differential image between the fully brine-saturated image and dry scan, including sub-resolution porosity. However, the oil saturation was not so straightforward to quantify at high Ca , since it was necessary to

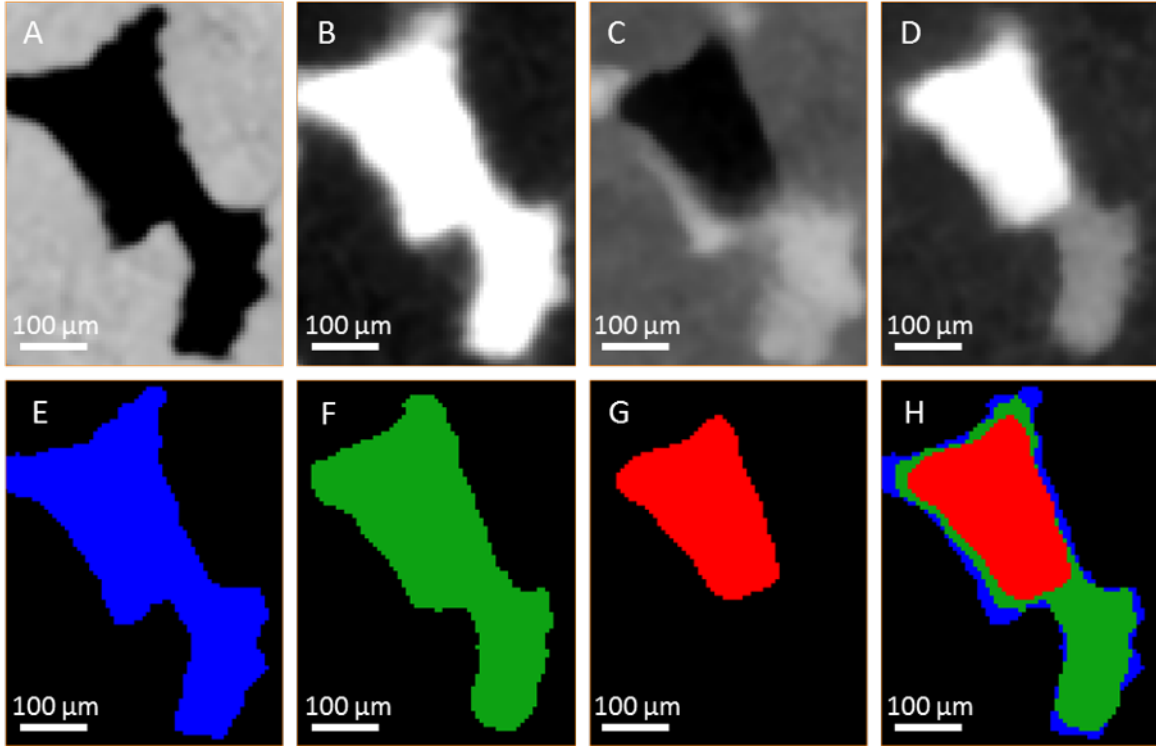


Figure 5.10: Illustration of the steps in segmentation: (A) An example pore of the dry scan. (B) The same pore imaged when fully brine-saturated. (C) The same pore imaged during steady-state multiphase flow at a fixed fractional flow. (D) Differential image between (B) and (C) after normalization. (E) The pore segmentation. (F) Segmented intermittent and oil phase in the pore space. (G) Segmented oil phase in the pore. (H) Segmented oil, brine and intermitty. Red is oil, blue is brine and green is the intermitty.

estimate the time-averaged oil saturation in the intermittent regions.

In order to identify the intermittent region, differential images between the brine scan and the two-phase scan at each fractional flow were obtained at first. As shown in Figure 5.10(D), the oil phase was highlighted in white, brine phase was black, and the intermittent region was in grey. Then a non-local means filter, which was introduced in Section 3.2.3, was applied to decrease the noise in the image. Later, the global thresholding method that was introduced in Section 3.2.4 was applied to segment the oil and intermitty based on their different grey-scale value ranges. Since the total pore space of the sample has been segmented based on the dry scan, the brine phase could be obtained by extracting the rest of the dry scan from brine and intermitty. Next, the segmented intermittent region, oil phase and brine phase were put as a mask onto the original raw image at each fractional flow to plot the grey scale distribution of each phase respectively as Figure 5.11(C) shows: we require these histograms to quantify the degree of intermitty. This method assumes that

the water saturation in the intermittent region is characterised by a distribution represented by average water saturation values for each phase. For each image different thresholds are chosen between peaks in the distribution of grey-scale values. As a sensitivity, different plausible thresholds were chosen – this did not affect the results significantly as the peaks were still identified clearly.

The grey-scale values where the peak values are located for each phase in the histogram are denoted as CT_{oil} for the stable oil phase, CT_{inter} for the intermittency, and CT_{brine} for the stable brine phase. These values represent the most likely CT numbers for each phase. In the multiphase images the contribution to the oil saturation in the intermittency was estimated as:

$$S_{oil,inter} = S_{inter}p_{oil} = S_{inter} \frac{CT_{brine} - CT_{inter}}{CT_{brine} - CT_{oil}} \quad (5.1)$$

where $S_{oil,inter}$ is the contribution to the oil saturation in the intermittency, S_{inter} is the fraction of the intermittency to the total pore volume, CT_{oil} is the average grey scale of the stable oil phase, CT_{inter} is the average grey-scale of the intermittency, and CT_{brine} is the average grey-scale of the brine phase. p_{oil} is the proportion of oil in the intermittency calculated from segmented images.

As shown in Figure 5.11, for the case when f_w is 0.5, the proportion of the oil in the intermittency, p_{oil} , is 0.308, while the fraction of the pore volume occupied by the intermittency, S_{inter} , is 0.328. Hence the contribution to the oil saturation in the intermittency $S_{oil,inter}$ is 0.101. The stable oil saturation is 0.339 and so the total oil saturation is 0.440.

5.2.5.3 Saturation at low and high Ca

Figure 5.12 shows the differential images at different fractional flows for the low Ca experiment: white indicates oil. In these experiments, the two fluid phases appear to have a temporally fixed pore-scale occupancy: this occupancy changes – there is less oil – only when the water fractional flow increases. Figure 5.13 shows the same images segmented into brine, oil and rock phases. Figure 5.14 shows the difference images for the high Ca experiment. Here, an intermediate grey phase is apparent, indicating that the occupancy in some of the pore space is intermittent. During the hour-long time of the scan it appears that some of the time these regions were oil-filled and at other times brine-filled. Figure 5.15 shows these images segmented into brine, oil, rock and intermittency.

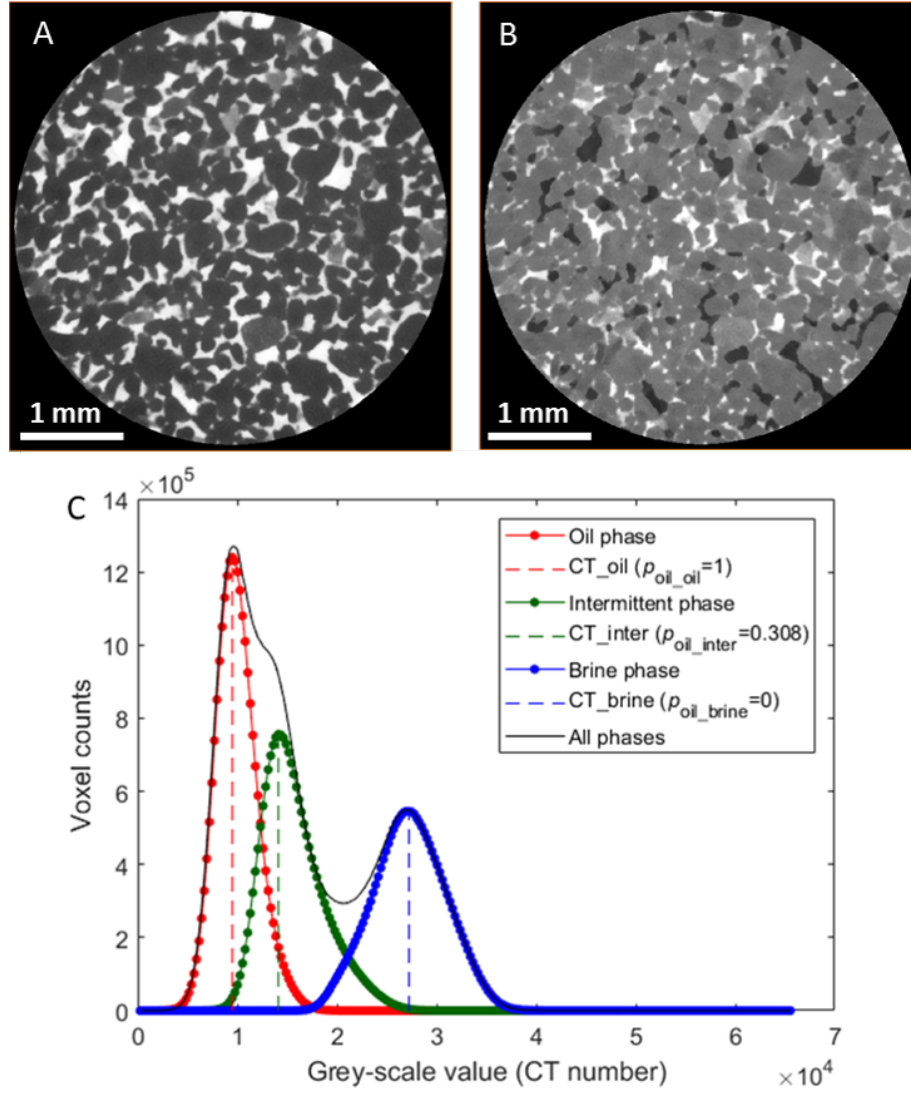


Figure 5.11: (A) A slice of the brine-saturated image. (B) A slice of multiphase image when f_w is 0.5 at high Ca . (C) The histograms for the three phases in the multiphase scan. The red dashed line indicates the average grey-scale of the oil phase, the blue dashed line indicates the average grey-scale of the brine phase, and the green dashed line shows the average grey-scale of the intermittency. The proportion of oil in the intermittency is 0.308. The contribution to the oil saturation in the intermittency is calculated using Equation 5.1.

In this case, the value is 0.101.

The average saturation of all phases, taken over the entire volume analysed, can be found, and the values are listed in Table 5.1. Overall the slice-averaged saturation is approximately constant along the imaged portion of the core, as shown in Figure 5.16.

For each of the segmented images at low and high Ca , we identified and labelled each unique oil cluster, as shown in Figure 5.17. Each cluster is given a different colour. The largest cluster has been coloured blue to help distinguish the connectivity. Clusters can either be

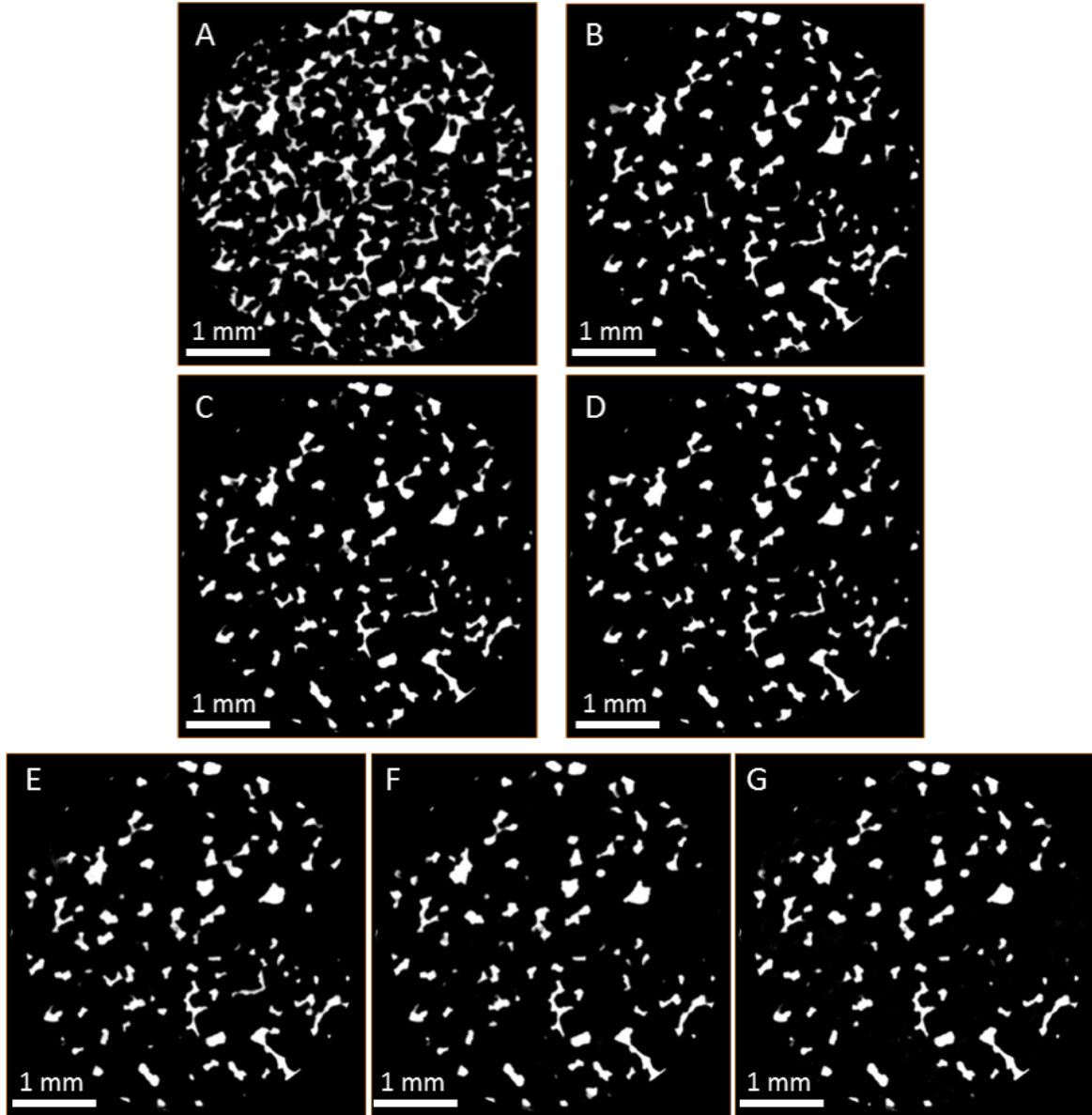


Figure 5.12: Two-dimensional cross-sections from three-dimensional differential images at different fractional flows when Ca is 3.0×10^{-7} . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Black includes grains and unchanged brine, and white is oil.

connected to the inlet, the outlet, or both, or be completely surrounded by other phases (these we call trapped ganglia).

Table 5.1 lists the proportion of oil, brine and intermittency and the fraction of the oil phase that connects from inlet to outlet, both including the intermittency and without it. When the fractional flow increases both at low and high Ca , the number of trapped oil ganglia increases and the volume proportion of the largest oil cluster to the whole oil volume decreases. At low Ca we see a rapid drop in connectivity at the end of imbibition

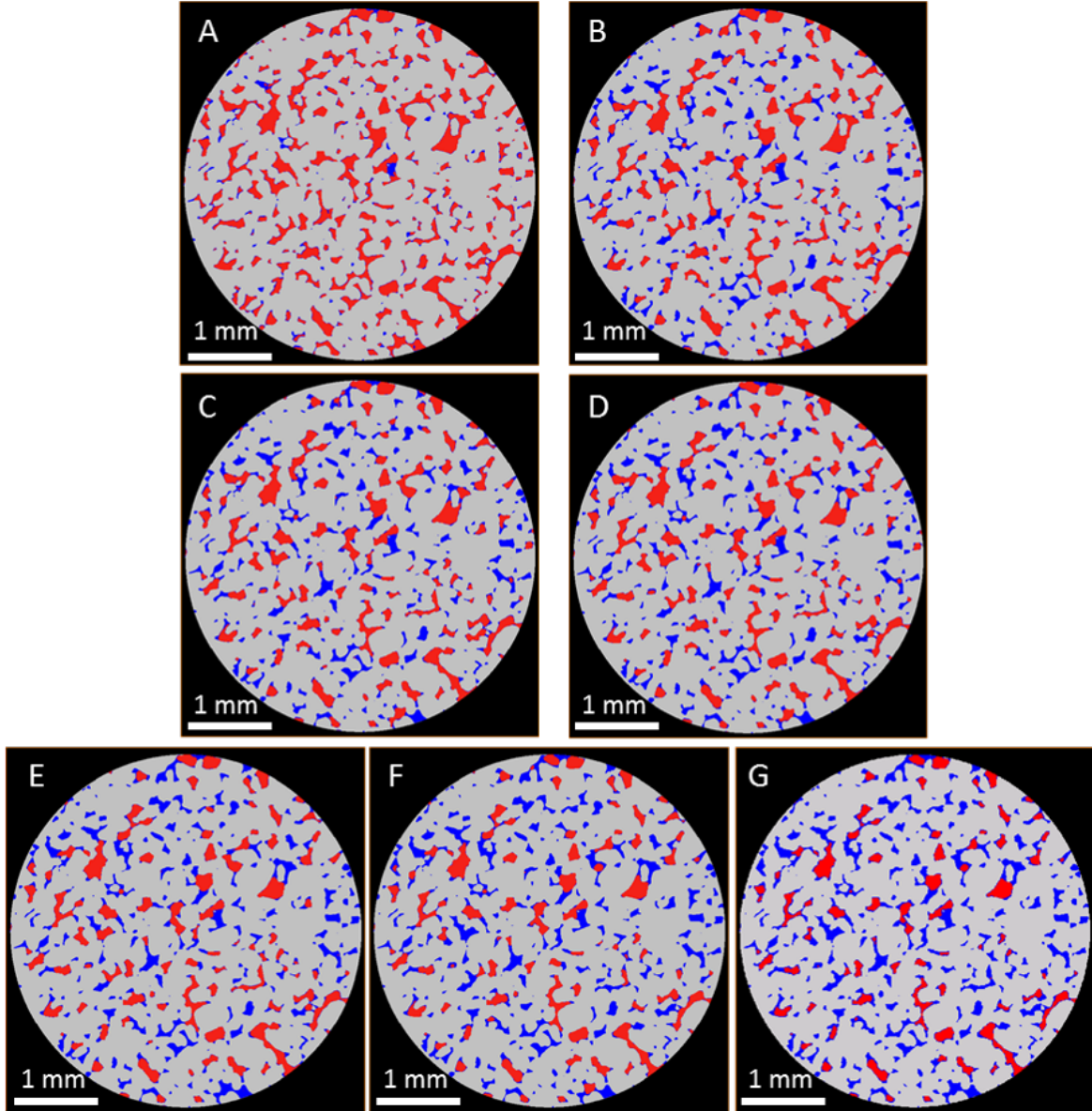


Figure 5.13: Two-dimensional cross-sections from three-dimensional segmented images at different fractional flows when Ca is 3.0×10^{-7} . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Grey is grain, red is oil and blue is brine.

and all the oil phase is trapped.

For the lower Ca the behaviour is consistent with the traditional Darcy model of multiphase displacement, where at steady state flow occurs through stable, connected sub-networks of each phase in the pore space (Blunt, 2017). For the higher Ca the behaviour is different: ignoring the intermittency, the oil is only connected at the highest oil saturations and yet can flow and continues to be displaced as the water fractional flow increases. Hence, connectivity allowing movement and displacement of the oil phase is only provided by the

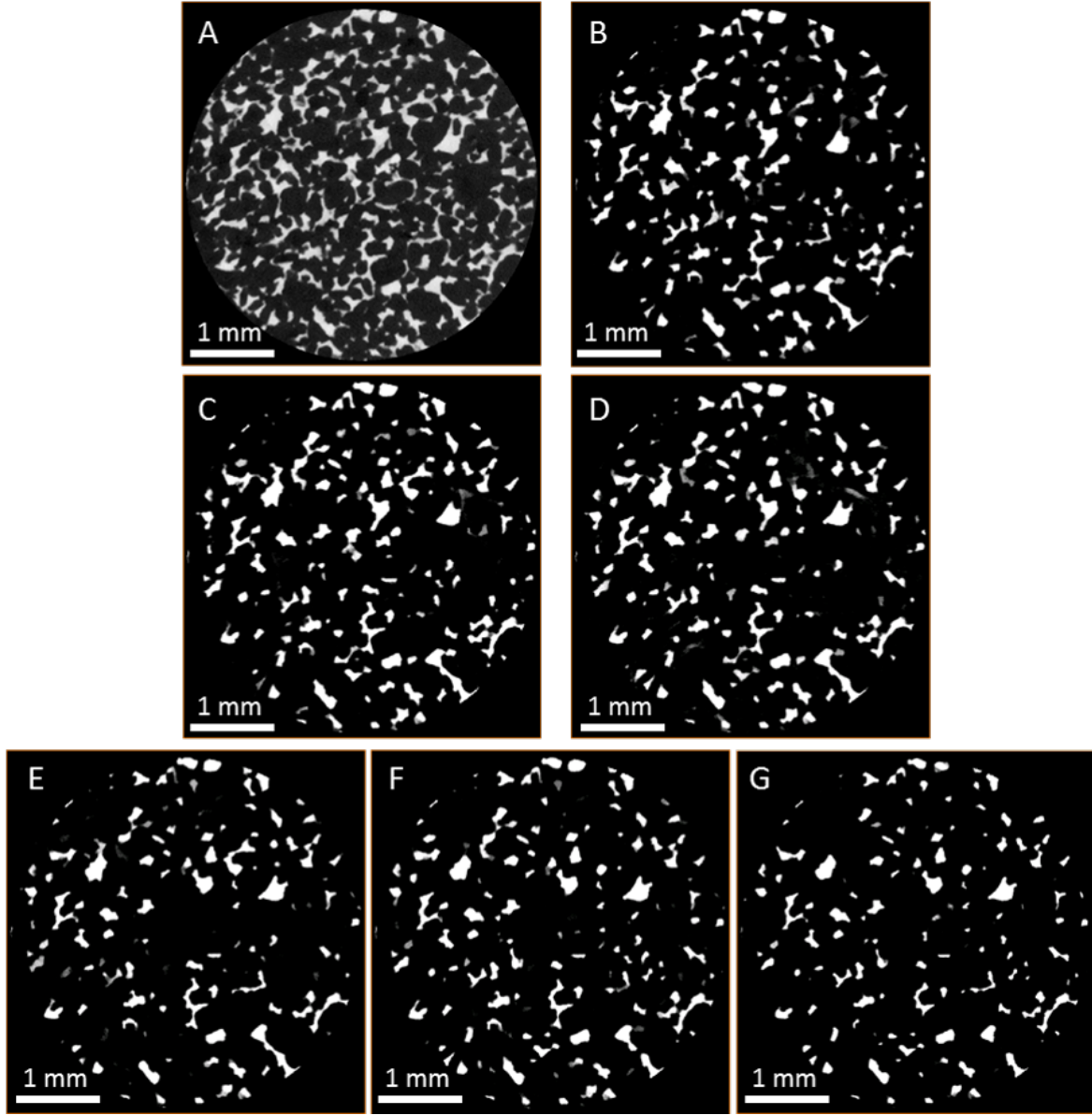


Figure 5.14: Two-dimensional cross-sections from three-dimensional differential images at different fractional flows when Ca is 7.5×10^{-6} . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Black includes grains and unchanged brine, white is oil and grey is the intermittency.

periodic occupancy of some of the pore space, as seen during fast imaging (Reynolds et al., 2017). However, in the work of Reynolds et al. (2017) fluctuations in pore occupancy and connectivity were observed even at the lowest flow rates or a Ca of 1.4×10^{-7} using the definition in this thesis: there are four possible reasons why this is not seen here. First, we consider a two-fluid system with a high non-wetting phase viscosity which may suppress pore-scale interfacial fluctuations, as opposed to the nitrogen-brine system studied by Reynolds et al. (2017). Second, Reynolds et al. (2017) considered only one fractional

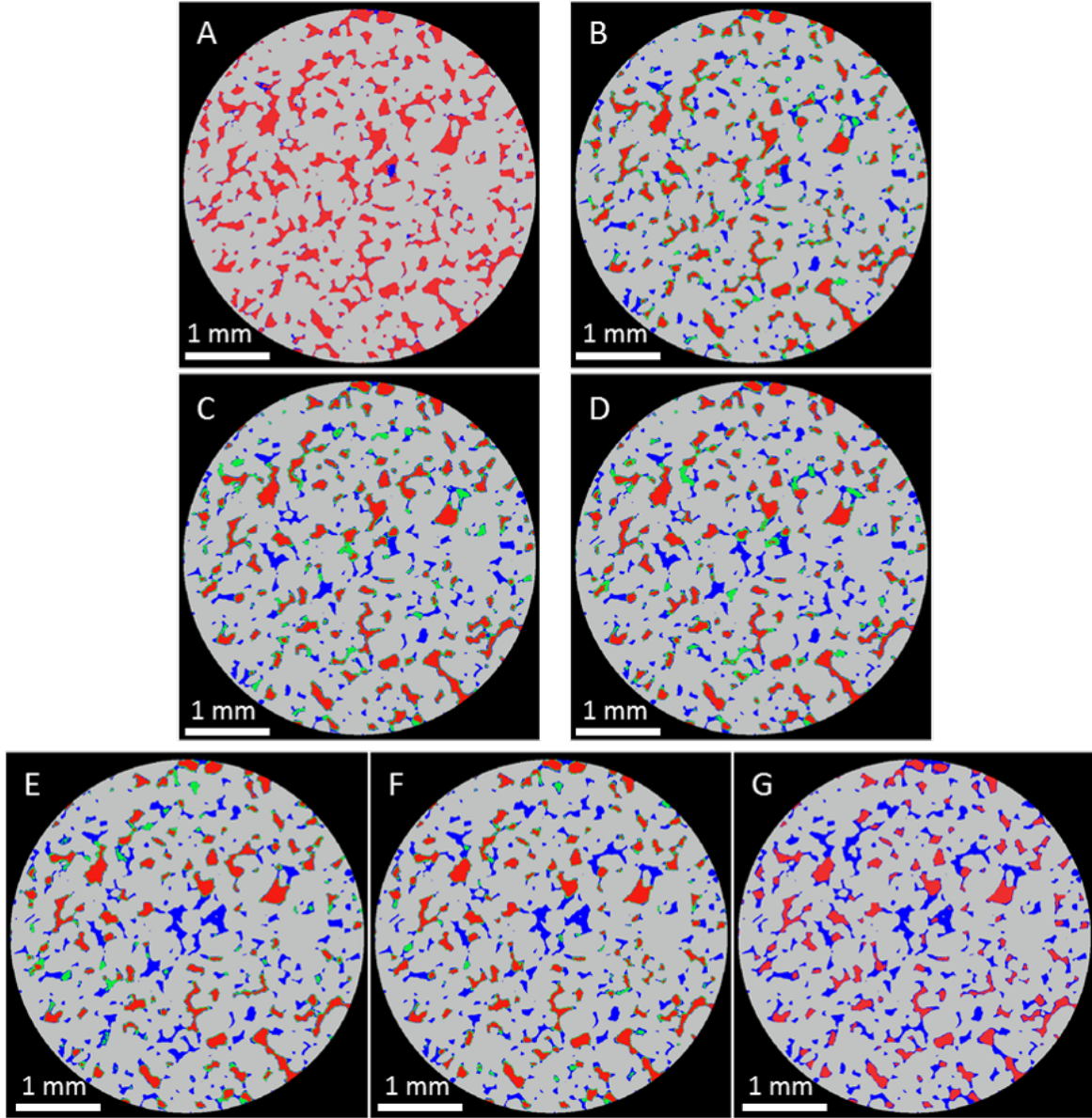


Figure 5.15: Two-dimensional cross-sections from three-dimensional segmented images at different fractional flows when Ca is 7.5×10^{-6} . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. The grey is grains, the red is oil, the blue is brine and the green is rearranged or intermittent oil phase.

flow during drainage where the nitrogen saturation was close to its percolation threshold, making disconnection possible with only a few displacement events. Third, Reynolds et al. (2017) injected the fluids for about 1 hour before scanning, whereas our work indicates that a longer time (around 20 hours) is needed to reach steady state. Finally, in our experiments we may be unable to distinguish regions of the void space where either water or oil only resides a small fraction of the time. Even accounting for the intermittency, we appear to have no oil connectivity for a water fractional flow of 0.85 in the high Ca experiment, which

5.2. Pore scale two-phase flow steady-state experiment in Bentheimer sandstone

Table 5.1: Percentages of the stable oil, stable water and intermittency, as well as the water saturation. Also shown is the fraction of the oil that connects from inlet to outlet, hence allowing flow and displacement, considering just the stable oil phase, and for the stable oil and intermittency combined.

f_w	$Ca = 3.0 \times 10^{-7}$		$Ca = 7.5 \times 10^{-6}$					
	S_w	Percentage of oil connected from inlet to outlet	Percentage of stable oil	Percentage of intermittency	Percentage of stable water	S_w	Percentage of oil and intermittency combined connected from the inlet to outlet	Percentage of stable oil connected from inlet to outlet
0	0.252	99.5	0.855	0	0.145	0.145	99.6	99.6
0.15	0.463	93.0	0.427	0.149	0.424	0.528	88.9	62.1
0.3	0.491	86.9	0.361	0.297	0.342	0.552	83.5	0
0.5	0.517	84.0	0.339	0.328	0.333	0.560	67.3	0
0.7	0.536	81.6	0.310	0.320	0.370	0.592	22.3	0
0.85	0.589	62.7	0.291	0.292	0.417	0.622	0	0
1	0.642	0	0.337	0	0.663	0.663	0	0

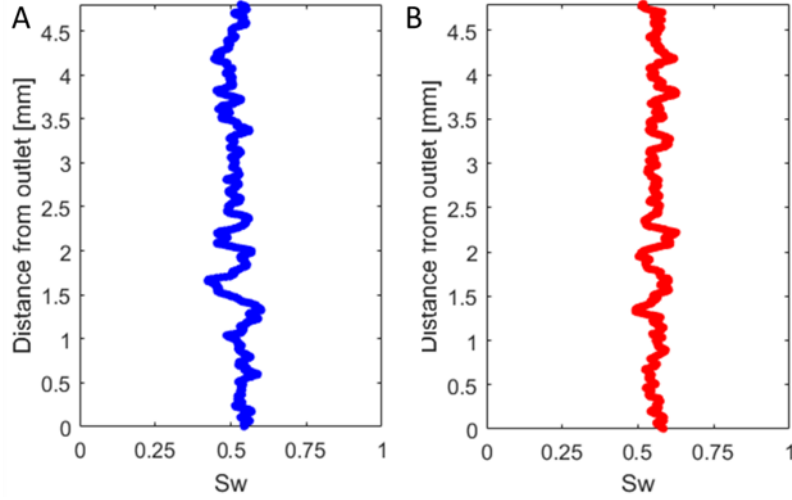


Figure 5.16: Sliced-averaged brine saturation distribution along the sample. (A) Brine saturation distribution when f_w is 0.5 at low Ca . The mean S_w is 0.517. (B) Brine saturation distribution when f_w is 0.5 at high Ca . The mean S_w is 0.560.

does suggest that we do not capture some pores that oil only occupies fleetingly.

The Euler characteristic (or number), is computed to indicate the connectivity of the non-wetting phase (Herring et al., 2013; Armstrong et al., 2016; Liu et al., 2017), and was calculated using the image analysis Avizo software. A larger negative value indicates that the oil is well connected. A positive value indicates poor connectivity, with more isolated

clusters than redundant loops in the structure. As shown in Figure 5.17, the non-wetting phase Euler characteristic was calculated at each fractional flow at each Ca (the second and fourth columns). In the third column, the Euler characteristic number is calculated by considering the oil and intermittency combined as the non-wetting phase. The Euler characteristic number increases with f_w , starting from a negative value and becoming positive, which shows that the non-wetting phase connectivity becomes lower as it is displaced by water. Even if we combine the intermittency and stable phase, the Euler characteristic is positive for all but the lowest water fractional flows. This indicates a poor connectivity of oil phase at high flow rates, with many isolated clusters. Again, the misidentification of some regions of the pore space occupied by one phase for a short time may lead us to underestimate the connectivity, since, as stated above, the oil continues to flow and is displaced.

5.2.6 Relative permeability

The pressure difference between the inlet and outlet of the core holder was measured using the pressure differential transducer shown in Figure 4.1. This was performed at steady-state conditions which were identified by observing a constant average saturation across the scanned volume, Figure 5.16, and a stable, constant pressure drop. In this experiment, we recorded the pressure drop during the last two hours of the total 10 - 20 hours during which a constant fractional flow was imposed. The scans were taken during the final hour. Figure 5.18 shows the pressure drop for each fractional flow together with the corresponding mean value and standard deviation during the final two hours: in all cases the results imply steady-state while the core was scanned: we have corrected for the additional pressure drop in the flow lines, see section 4.1.2.

For $f_w = 0$ and 1, and for low Ca , the pressure profile is smooth, which indicates an unchanging fluid configuration, consistent with the absence of an intermittency in these cases. However, for intermediate f_w and high Ca , the rapid fluctuations in pressure are measured (Rücker et al., 2015a; Schlüter et al., 2017; Sinha et al., 2017), which are consistent with intermittency. The hypothesis is that these represent local pore-scale displacement events, rather than being an artefact of the pump rates or noise in the transducers.

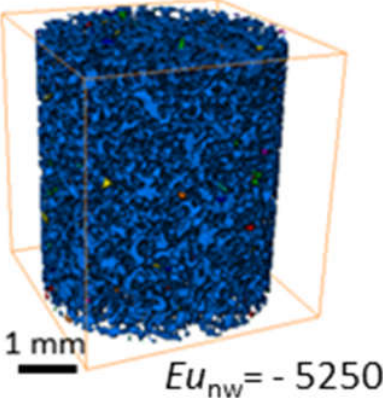
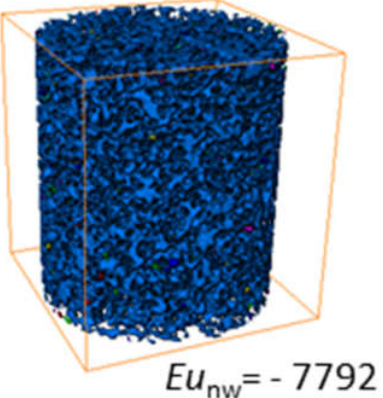
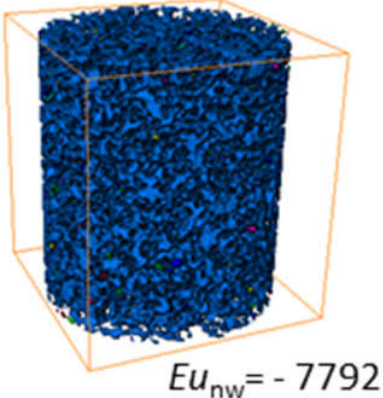
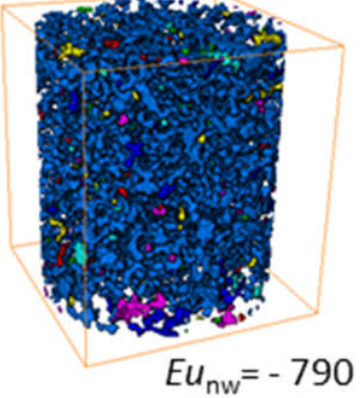
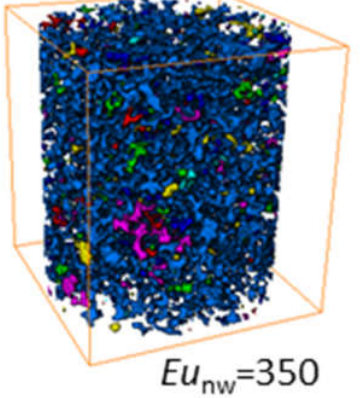
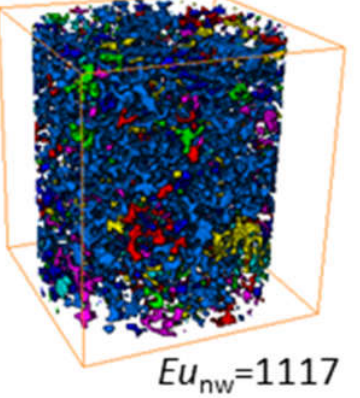
f_w	$Ca=3.0 \times 10^{-7}$	$Ca=7.5 \times 10^{-6}$	
	Oil clusters	Clusters of oil and intermittency combined	Oil clusters excluding the intermittency
0	 $Eu_{nw} = -5250$	 $Eu_{nw} = -7792$	 $Eu_{nw} = -7792$
0.15	 $Eu_{nw} = -790$	 $Eu_{nw} = 350$	 $Eu_{nw} = 1117$

Figure 5.17: Three-dimensional images of oil clusters at different fractional flows when Ca is 3.0×10^{-7} and 7.5×10^{-6} . Each cluster is given a different colour. Blue shows the largest oil cluster. The second column shows oil clusters at low Ca . The third column shows clusters of the oil and intermittent phases combined at high Ca , and the fourth column shows clusters of just the oil, excluding the intermittency. The number in the second and the fourth column is the non-wetting phase Euler characteristic, Eu_{nw} . The number in the third column is the Euler characteristic of stable oil and intermittent phases combined.

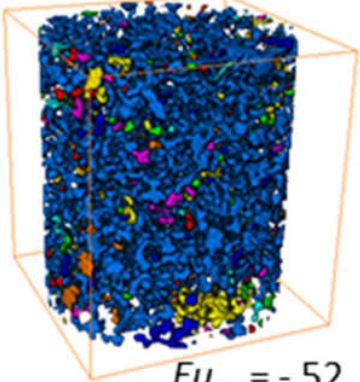
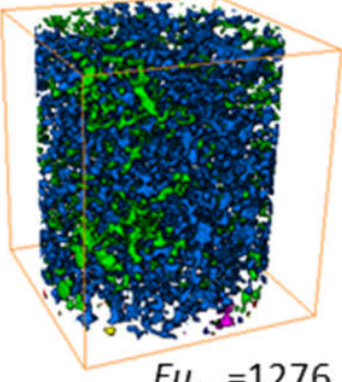
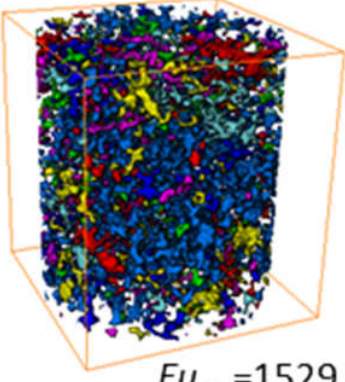
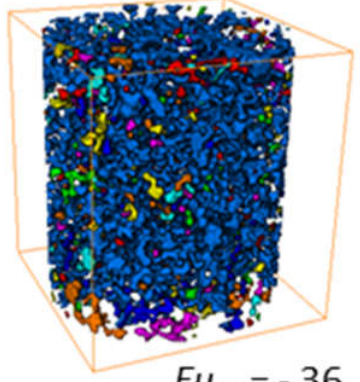
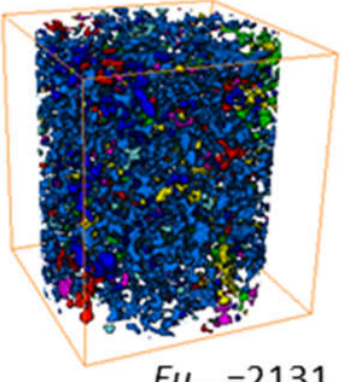
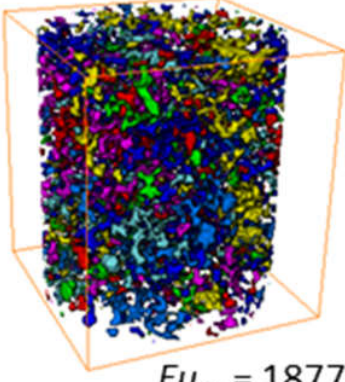
f_w	$Ca=3.0 \times 10^{-7}$	$Ca=7.5 \times 10^{-6}$	
	Oil clusters	Clusters of oil and intermittency combined	Oil clusters excluding the intermittency
0.3	 $Eu_{nw} = -52$	 $Eu_{nw} = 1276$	 $Eu_{nw} = 1529$
0.5	 $Eu_{nw} = -36$	 $Eu_{nw} = 2131$	 $Eu_{nw} = 1877$

Figure 5.17: (continued). Three-dimensional images of oil clusters at different fractional flows when Ca is 3.0×10^{-7} and 7.5×10^{-6} . Each cluster is given a different colour. Blue shows the largest oil cluster. The second column shows oil clusters at low Ca . The third column shows clusters of the oil and intermittent phases combined at high Ca , and the fourth column shows clusters of just the oil, excluding the intermittency. The number in the second and the fourth column is the non-wetting phase Euler characteristic, Eu_{nw} . The number in the third column is the Euler characteristic of stable oil and intermittent phases combined.

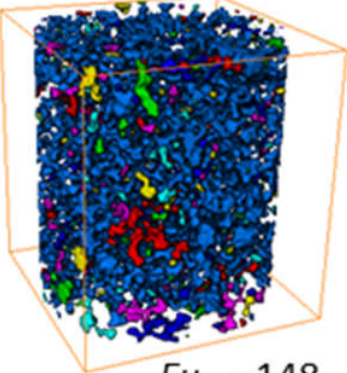
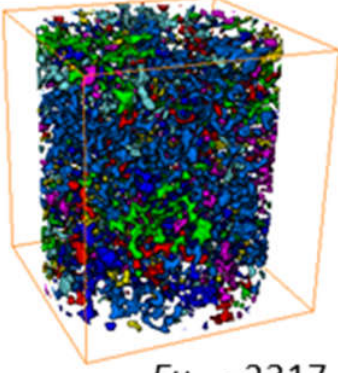
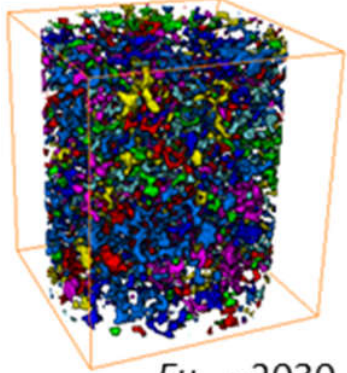
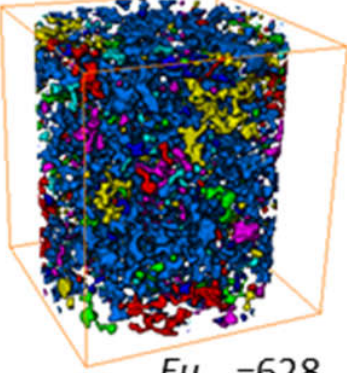
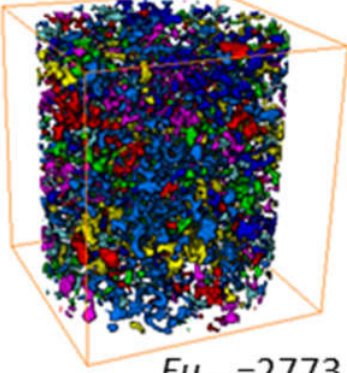
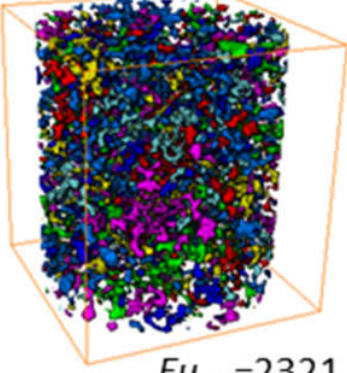
f_w	$Ca=3.0 \times 10^{-7}$	$Ca=7.5 \times 10^{-6}$	
	Oil clusters	Clusters of oil and intermittency combined	Oil clusters excluding the intermittency
0.7	 $Eu_{nw}=148$	 $Eu_{nw}=2317$	 $Eu_{nw}=2030$
0.85	 $Eu_{nw}=628$	 $Eu_{nw}=2773$	 $Eu_{nw}=2321$

Figure 5.17: (continued). Three-dimensional images of oil clusters at different fractional flows when Ca is 3.0×10^{-7} and 7.5×10^{-6} . Each cluster is given a different colour. Blue shows the largest oil cluster. The second column shows oil clusters at low Ca . The third column shows clusters of the oil and intermittent phases combined at high Ca , and the fourth column shows clusters of just the oil, excluding the intermittency. The number in the second and the fourth column is the non-wetting phase Euler characteristic, Eu_{nw} .

The number in the third column is the Euler characteristic of stable oil and intermittent phases combined.

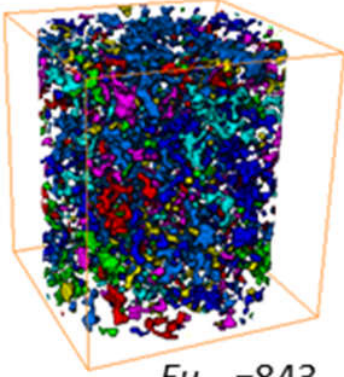
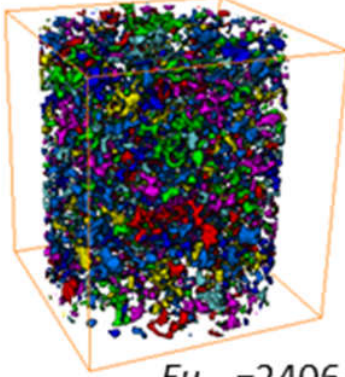
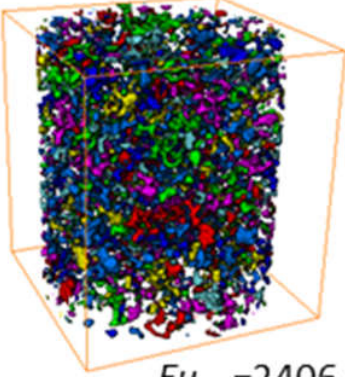
f_w	$Ca=3.0 \times 10^{-7}$	$Ca=7.5 \times 10^{-6}$	
	Oil clusters	Clusters of oil and intermittency combined	Oil clusters excluding the intermittency
1	 $Eu_{nw}=843$	 $Eu_{nw}=2406$	 $Eu_{nw}=2406$

Figure 5.17: (continued). Three-dimensional images of oil clusters at different fractional flows when Ca is 3.0×10^{-7} and 7.5×10^{-6} . Each cluster is given a different colour. Blue shows the largest oil cluster. The second column shows oil clusters at low Ca . The third column shows clusters of the oil and intermittent phases combined at high Ca , and the fourth column shows clusters of just the oil, excluding the intermittency. The number in the second and the fourth column is the non-wetting phase Euler characteristic, Eu_{nw} .

The number in the third column is the Euler characteristic of stable oil and intermittent phases combined.

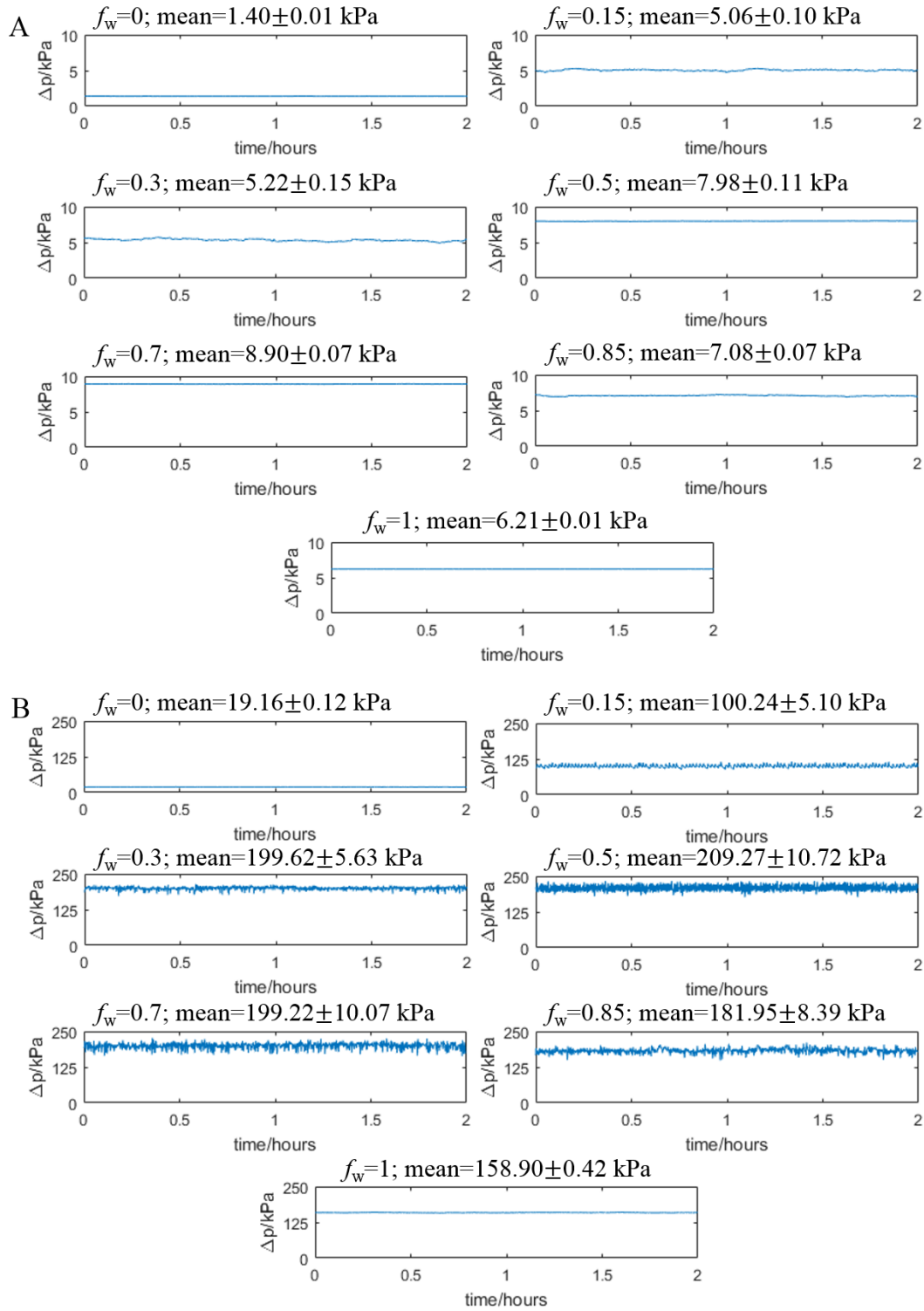


Figure 5.18: (A) Pressure difference measured between the inlet and outlet of the samples at low Ca . (B) Pressure difference measured between the inlet and outlet of the samples at high Ca . The mean pressure drop and standard deviation at each fractional flow is shown.

A typical throat radius, r , for Bentheimer is $24 \mu\text{m}$ (Blunt, 2017); a representative capillary

pressure for a piston-like event is $2\sigma/r$, or around 4 kPa in this case. At low Ca , the standard deviation in the pressure drop is much lower than this, meaning that fluid re-arrangement is limited, whereas for higher Ca , the pressure fluctuations are sufficiently large to allow local displacement in the pore space. This indicates that the pressure fluctuations are consistent with intermittency. The energy supplied from pumping fluids through the core is sufficient to allow a significant fraction of the pore space to experience displacement at the higher capillary number (Reynolds et al., 2017).

The absolute permeability measured using the same transducer is $2.045(\pm 0.056) \times 10^{-12}$ m². The relative permeability is calculated by:

$$k_{rw} = \frac{Q_w \mu_w L}{A \Delta P k}; k_{ro} = \frac{Q_o \mu_o L}{A \Delta P k} \quad (5.2)$$

where k_{rw} is brine relative permeability, k_{ro} is oil relative permeability, Q_w is brine flow rate, m³/s, Q_o is oil flow rate, m³/s, μ_w is brine viscosity, Pa·s, μ_o is oil viscosity, Pa·s, L is the length of the whole sample, m, A is the cross area of the sample, m², k is absolute permeability, m², and ΔP is the pressure difference across the sample, Pa.

Table 5.2: Saturation and relative permeability calculated at low and high Ca over a range of f_w .

f_w	$Ca = 3.0 \times 10^{-7}$			$Ca = 7.5 \times 10^{-6}$		
	S_w	k_{ro}	k_{rw}	S_w	k_{ro}	k_{rw}
0	0.252	0.310	0.000	0.145	0.567	0.000
0.15	0.463	0.073	0.013	0.528	0.092	0.015
0.3	0.491	0.058	0.025	0.552	0.038	0.016
0.5	0.517	0.027	0.027	0.560	0.026	0.025
0.7	0.536	0.015	0.034	0.592	0.016	0.037
0.85	0.589	0.009	0.051	0.622	0.009	0.050
1	0.642	0.000	0.069	0.663	0.000	0.067

Table 5.2 lists the wetting phase saturation with the measured oil and brine relative permeabilities at each fractional flow for low and high capillary numbers. Note that the saturation values are estimated from only the scanned region of the core, while the pressure drops are measured across the whole sample. Figure 5.19 shows the estimated fractional flow curves at steady state for water-oil imbibition at low and high capillary numbers. Table 5.2 and Figure 5.20 shows the relative permeabilities plotted as functions of wetting-phase saturation. At the lower Ca , the initial water saturation at the end of primary drainage was 0.252 ± 0.030 and the residual oil saturation at the end of imbibition was 0.358 ± 0.005 .

The errors are estimated from the root-mean square uncertainties in phase volume, total volume, and porosity during image segmentation. At the higher Ca , the initial water saturation and residual oil saturation were 0.145 ± 0.031 and 0.337 ± 0.003 , respectively. Note that at the higher flow rate we see finite relative permeabilities even where regions of the pore space always occupied by oil (for a given fractional flow) do not connect from inlet to outlet. The only way that the oil can be displaced is through the intermittent filling of some pores allowing portions of the oil to be connected and flow.

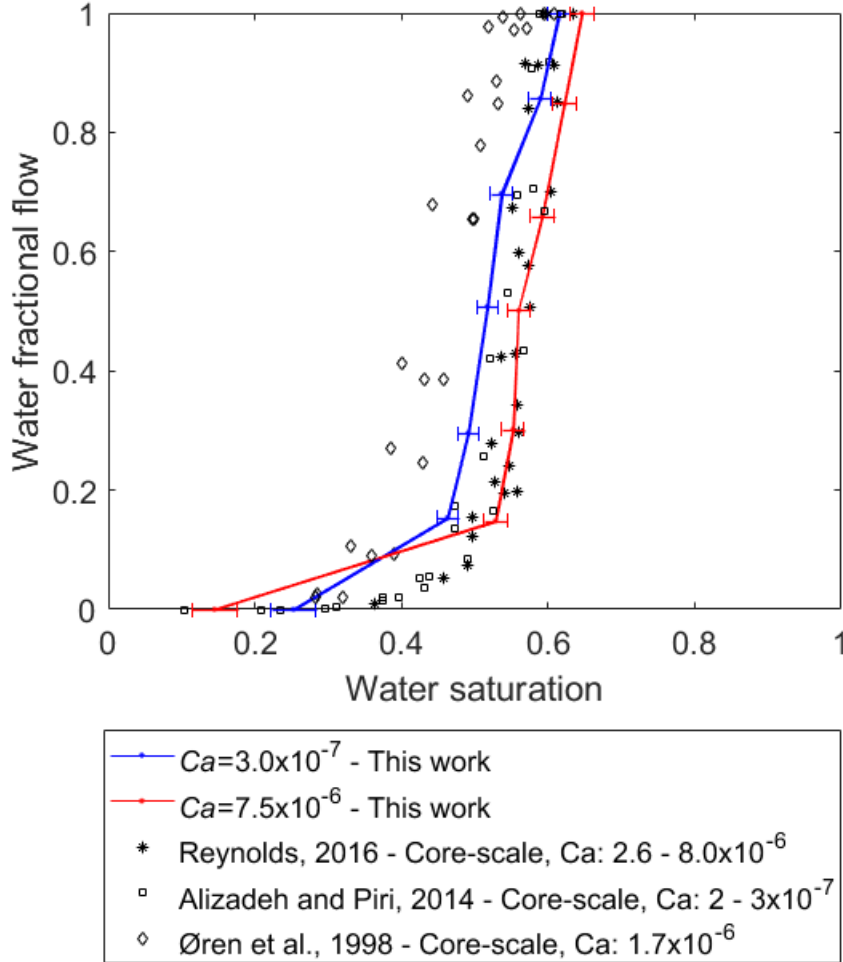


Figure 5.19: Fractional flow curves calculated at steady state for water-oil imbibition at low and high capillary number, $f_w(S_w)$ shown by the blue and red lines respectively. The results are compared to five sets of cm-scale experimental data by Reynolds (2016), three sets of cm-scale experimental data by Alizadeh and Piri (2014), and three sets of cm-scale experimental data by Øren et al. (1998). Error bars in the x-direction consider uncertainties in the measured saturation caused by the threshold value chosen during image segmentation.

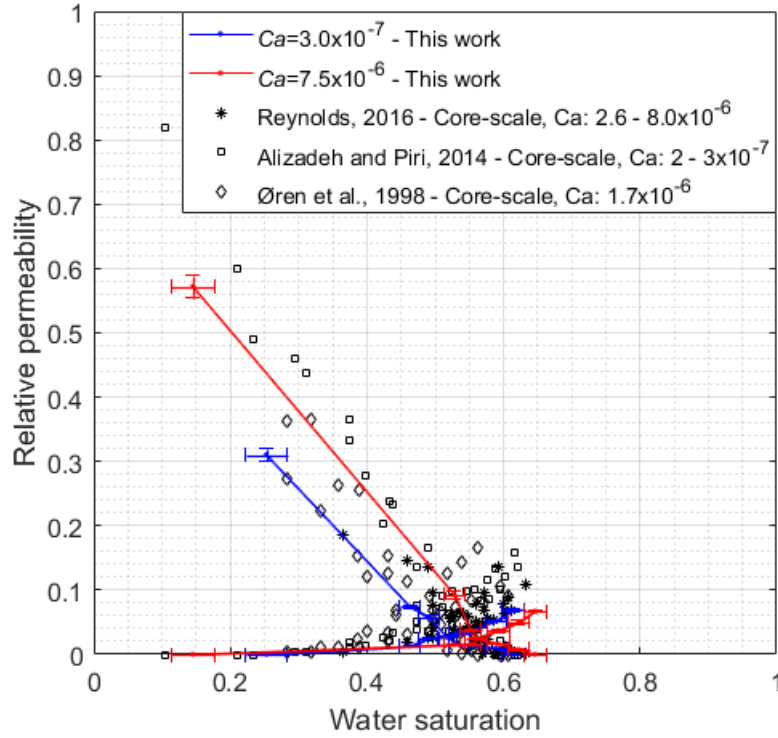


Figure 5.20: Relative permeabilities calculated at steady state for water-oil imbibition at low and high capillary number, Ca shown by the blue and red lines respectively. The results are compared to five sets of cm-scale experimental data by Reynolds (2016), three sets of cm-scale experimental data by Alizadeh and Piri (2014), and three sets of cm-scale experimental data by Øren et al. (1998). Error bars in the y-direction consider uncertainties in the measurement of the length and diameter of the sample, the absolute permeability, pressure measurements and the pump rates. Error bars in the x-direction consider uncertainties in the measured saturation caused by the threshold value chosen during image segmentation.

The relative permeability data and the fractional flow, $f_w(S_w)$, obtained by our experiment at the mm-scale are compared with the data collected from experiments in the literature which were conducted at the core scale, as shown in Table 5.3.

Table 5.3: Comparison of Ca and sample size between this work and published work.

Author	Fluid system	Diameter/mm	Length/mm	Ca
This work	Oil-water	4.99 ± 0.01	60 ± 0.01	3.0×10^{-7} ; 7.5×10^{-6}
Reynolds (2016)	CO ₂ -water; N ₂ -water	38	239	$2.6 - 8.0 \times 10^{-6}$
Alizadeh and Piri (2014)	Oil-water; Gas-water; Gas-oil	38.1	152.4	$2 - 3 \times 10^{-7}$
Øren et al. (1997)	Oil-water	-	-	1.7×10^{-6}

In this work we used cores that were smaller than those used in conventional analysis. Our results are only valid if our sample size exceeds the representative elementary volume. Arns et al. (2003) demonstrated numerically that cubes of size 1 mm^3 or more are sufficiently large to estimate permeability for the Bentheimer sandstone used in this study. Hilfer and Lemmer (2015) found that for Fontainebleau sandstone, the representative elementary volume is around 3 mm (a volume of 27 mm^3) for porosity and around 250 mm^3 for permeability. Bentheimer is a homogeneous sandstone with a grain size from $0.1 - 0.38 \text{ mm}$ (Reynolds and Krevor, 2015), which is similar to Fontainebleau sandstone which has a grain size from $0.1 - 0.4 \text{ mm}$ (Delfosse-Ribay et al., 2004). For saturation, our imaging volume is 68 mm^3 , while the total sample volume (used for the pressure measurements) is 1180 mm^3 ; this latter volume is above the representative elementary volume calculated by Hilfer and Lemmer (2015), while the constancy of average saturation within the imaging volume, Figure 5.16, indicates that this is also accurately estimated.

As Figure 5.20 shows, our data is within the range of larger-scale experiments, with the low Ca results closer to the low Ca experiments of Alizadeh and Piri (2014) and the high Ca results closer to the higher Ca experiments of Reynolds (2016). In this case, for a homogeneous sandstone, we suggest that we are able, simultaneously, to assess pore-scale occupancy and measure macroscopic properties, such as saturation and relative permeability, reliably.

5.2.7 Summary

We have developed an X-ray tomography based experimental and image analysis methodology to measure the relative permeability and associated pore-scale pore occupancy. We have performed imbibition using oil-brine and measured steady-state relative permeability on a sandstone rock core at low and high capillary number, Ca : 3.0×10^{-7} and 7.5×10^{-6} . During the hour-long scan time it appeared that there were regions of the pore space that were occupied alternately by oil and brine. This intermittent flow was identified by obtaining the differential image between a doped brine image and the scans taken at each fixed fractional flow. The pressure drops at each fractional flow at low and high Ca were measured by high-precision pressure differential sensors and used to calculate the relative permeability.

We show that at low Ca , the oil and water phase occupancy appears to be constant with

flow provided by fixed connected sub-networks through the pore space, consistent with conventional theory. At higher Ca , we see that a significant fraction of the pore space is occupied by this intermittency, interpreted as regions of the void space where, during the scan, oil and water are both present. The oil phase in regions where it is always present is not connected, and yet it continues to flow and is displaced when the water fractional flow increases. The intermittent regions of the pore space are therefore necessary to allow the oil to flow, even if the connectivity is only periodically present.

The results from our mm-scale experiment demonstrate how two fluids flow at the microscopic level. The measured relative permeabilities are compared with literature measurements on larger cm-scale cores. Overall, our data is within the range and close to the core-scale steady state data published by Reynolds (2016), whose samples were taken from the same block as ours. Since Bentheimer is strongly water-wet, in a homogeneous unsteady state one-dimensional displacement, our results would imply for the same fluid viscosities a piston-like macroscopic displacement with a shock from S_{wi} to $1 - S_{or}$. This has been seen for other strongly water-wet media (Blunt, 2017).

In future work, we could repeat these experiments, but at a synchrotron imaging facility with faster scan times to confirm, directly, this intermittent or dynamic connectivity, as seen already for brine-nitrogen flow (Reynolds et al., 2017): these results are presented in Chapter 6. The relative permeability could also be computed from the measured pore-scale configurations of fluid and compared to the experimental measurements (Armstrong et al., 2016). In addition, the technique could be applied to a wide range of rock types and wettabilities: indeed, the extension to a micro-porous carbonate is presented next. Lastly, the quantitative analysis of pore occupancy could be used for validation or input into simulation studies to explain and predict the observed displacement behaviour.

5.3 Pore scale two-phase flow steady state experiment in Estailades carbonate

This work has been published in Gao et al. (2019).

5.3.1 Introduction

Carbonates are known to have a complex pore space whose heterogeneous structure and connectivity have a significant impact on flow, transport and reactive properties (Gouze et al., 2008; Bijeljic et al., 2013a; Apourvari and Arns, 2016). In particular, micro-porosity, while it may control the flow and transport behaviour, is challenging to describe using current imaging and modelling methods (Ghous et al., 2007; Tian and Wang, 2018).

Recently, an improved understanding of the role of micro-porosity in single-phase flow in carbonates (Lin et al., 2016; Bijeljic et al., 2018) and two-phase flow in a dual porosity sandstone (Lin et al., 2017) has been provided using differential imaging micro-tomography. Importantly, the differential imaging method provides information on additional connectivity between macro-pores through micro-porosity. The quantification of micro-porosity can then be used in modelling studies (Tian and Wang, 2018). In Section 5.2, differential imaging was used during steady-state two-phase flow in Bentheimer sandstone (Gao et al., 2017), in which an intermittent flow was distinguished in the macroscopic pore space which was periodically occupied by either oil or brine. This method helps to recognize the fast fluid behaviours during the comparatively slow micro-CT scan. In this section we will combine these two applications of differential imaging to study two-phase flow in a micro-porous carbonate, for which the pore space is more complex.

In this section, we examine pore occupancy in two-phase flow in the complex pore space of Estailades carbonate, with significant sub-resolution micro-porosity. Our goal is to examine the role of micro-porosity on fluid distribution and relative permeability, measured directly from the pressure differential at different fractional flows (the ratio of brine flow rate to the total flow rate of oil and brine) during a steady-state experiment.

The sample used here is Estailades carbonate, which contains approximately 97.9% calcite and 2.1% quartz (analysis conducted at Weatherford Laboratories, East Grinstead, UK). The sample, called a core, was a cylinder 6.01 ± 0.01 mm in diameter and 51.34 ± 0.01 mm in length. The absolute permeability of the same sample was found from Darcy's law and the pressure differential measured at two flow rates, 0.2 mL/min and 0.5 mL/min, to be $1.06(\pm 0.02) \times 10^{-13}$ m². The total porosity, estimated from the micro-CT image, is 28.4(± 0.5)% out of which, micro-porosity accounted for 21.5%, while the macro porosity was 6.9%. The helium porosity of the same sample was 30.3(± 0.2)%; the image-based value

is slightly lower, indicating that some micro-porosity was not correctly captured.

The non-wetting phase was decane that has a density of 730 kg/m^3 (n-Decane, Acros Organics) and a viscosity of $0.838 \text{ mPa}\cdot\text{s}$ (provided by PubChem, open chemistry database). To provide an optimal contrast between brine, oil and rock phases, 30 wt% potassium iodide (KI) brine was used as the wetting, aqueous, phase. The density of brine was measured to be $1263 \pm 2 \text{ kg/m}^3$ at ambient conditions by weighing a 1 mL drop of the liquid. The viscosity of the brine was $0.82 \pm 0.01 \text{ mPa}\cdot\text{s}$. Using the pendant drop method (Andreas et al., 1938; Stauffer, 1965), the interfacial tension between brine and decane was measured to be $47 \pm 2 \text{ mN/m}$ at ambient conditions by a Ramé-Hart apparatus (590 F4 series).

The total flow rate used in this experiment was 0.7 mL/min . This was kept fixed during the whole experiment although fractional flow was changed. The capillary number was calculated from the total flow rate. The viscosity used in the calculation of capillary number was the mean value of oil and brine viscosities. Thus the capillary number for this experiment was 7.3×10^{-6} . The fractional flow was defined as the volumetric injection rate of the aqueous phase divided by the total fluid rate of both the oil and aqueous phases: $f_w = q_w/q_t$, where q_w is the aqueous (brine) phase Darcy velocity and q_t is the total velocity. The capillary number is at the upper end of that encountered in reservoir settings and was chosen so that some intermittent flow would be observed, based on our previous work in a sandstone (Gao et al., 2017).

The fluid configurations were imaged using a Zeiss XRM-510 X-ray microscope at a voxel size of $3.58 \text{ }\mu\text{m}$ using a flat panel detector. The X-ray energy was 75 keV , the exposure time was 0.5 s and the number of projections was 1601. All these projections were reconstructed into a three-dimensional image using proprietary software on the Versa system. Two images were taken and then stitched together to obtain a larger field of view to analyse. Then the stitched images were cropped into cylindrical sections to avoid the sleeve being shown at the boundaries of the core. The stitched image size was $1582 \times 1582 \times 2789$ voxels, 5.66 mm in diameter and 9.98 mm in length, which represents a bulk volume of 251.1 mm^3 . All reconstructed images were registered to the dry scan image to have the same orientation, and then Lanczos algorithm was used to resample them (Burger and Burge, 2016).

We first characterise the pore space in Section 5.3.2. Then in Sections 5.3.3 and 5.3.4 we quantify the fluid occupancy in the micro- and macro-pore space. Furthermore, in Section

5.3.5 we demonstrates change in fluid occupancy with fractional flow, while the associated steady-state relative permeability measurements are presented in Section 5.3.6. In Section 5.3.7 we quantify and examine the intermittent regions dependence on pore and throat size. In Section 5.3.8, we analyse the ganglia size distributions for each fractional flow.

5.3.2 Characterization of macro- and micro-pore space

In contrast to the earlier studies on sandstones, presented in the previous section, Estailades carbonate has a bimodal pore structure with inter-granular macro-porosity and micro-porosity, whose pore throat radius distribution clearly shows two peaks around 0.2 and 10 μm from mercury injection capillary pressure analysis, see Figures 5.5 and 5.6 (Bijeljic et al., 2013a; Alyafei and Blunt, 2016), and as indicated in the cross-sectional images in Figure 5.21. Figure 5.21a and Figure 5.21b show images of the dry scan and the scan of the sample saturated with 30 wt% KI brine, respectively. We have designed the experiment so that brine has the strongest X-ray adsorption – even greater than solid – and appears bright (white) in the raw images, as with the sandstone. Using the differential imaging method, we subtract the dry scan from the brine scan to distinguish micro-porosity from macroscopic pore space. Figure 5.21c is the difference image between Figure 5.21b and Figure 5.21a, from which solid grain, macro-pore space and micro-porosity can be discriminated. A non-local means filter was applied to the differential images to reduce the noise while preserving edges (Buades et al., 2005; Buades et al., 2008). Then global thresholding was used to segment the image into different phases (Lin et al., 2016). The details of the image processing are provided in Chapter 3.

The histogram of the grey-scale CT numbers for the dry scan is presented in Figure 5.22. Here solid grain, micro-porous and macro-pore space can be distinguished. Fluid occupancy in micro-pore and macro-pore space is studied in Sections 5.3.3 and 5.3.4, respectively.

5.3.3 Fluid occupancy and porosity in the micro-pore space

Grey-scale images are shown in Figure 5.23, where two-dimensional cross-sections of three-dimensional raw micro-CT images at different fractional flows are presented. The grey

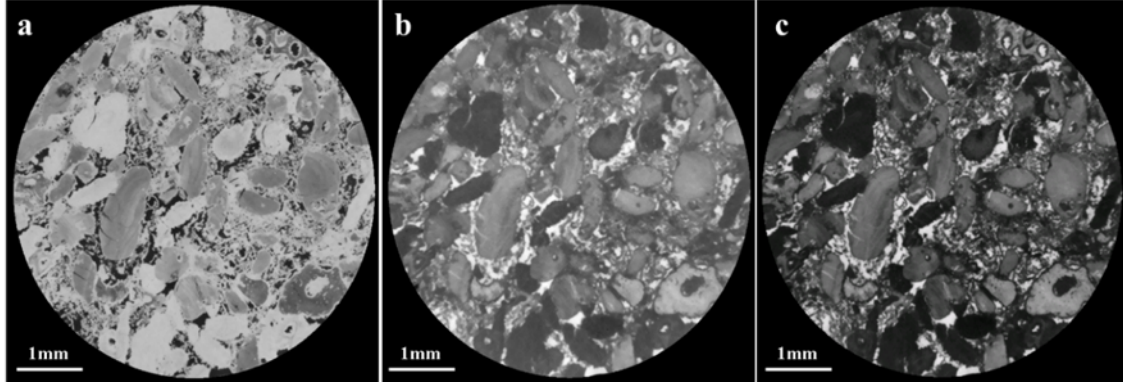


Figure 5.21: Two-dimensional cross-sections of three-dimensional micro-CT images at the same location in the Estailades sample. (a) The dry scan. (b) The brine-saturated image. (c) The difference image between (b) and (a). Black in (c) represents the impermeable solid grains, white is macro-pore space, while intermediate grey values indicate micro-porosity where the pore structure cannot be explicitly resolved in the image.

represents grains and intermittent regions in macro-pores, as well as micro-porosity, black is stable oil in macro-pores, while white is brine in macro-pores.

The first observation is that there are some voxels in the micro-porous space which did not change grey-scale value for all fractional flows, indicating that they were always filled with brine. The second observation is that, with increasing fractional flow, some voxels in the micro-porous space significantly increased their grey-scale value which became similar to the value of brine-only voxels; these are the regions which are indicated by the red circles. This shows that oil in the micro-porosity was displaced by brine.

Differential imaging can be used to discriminate the fluid occupancy in micro-porosity – separating the voxels filled only with brine, and the voxels that were partially filled by brine and oil. This fluid occupancy was then determined by calculating the difference image between the fully brine-saturated scan and the scan at each fractional flow.

Image normalization was then used to ensure that the grey-scale value of each voxel was the same for the images taken at different fractional flows if the fluid occupancy was unchanged. Thus the peak values of the grey-scale histogram distribution for multiphase images were rescaled to be the same as those of the brine saturated image. Then the difference image between the brine scan and the multiphase flow scan is defined as:

$$CT(difference) = CT(brine\ scan) - CT(multiphase\ scan) + CT0 \quad (5.3)$$

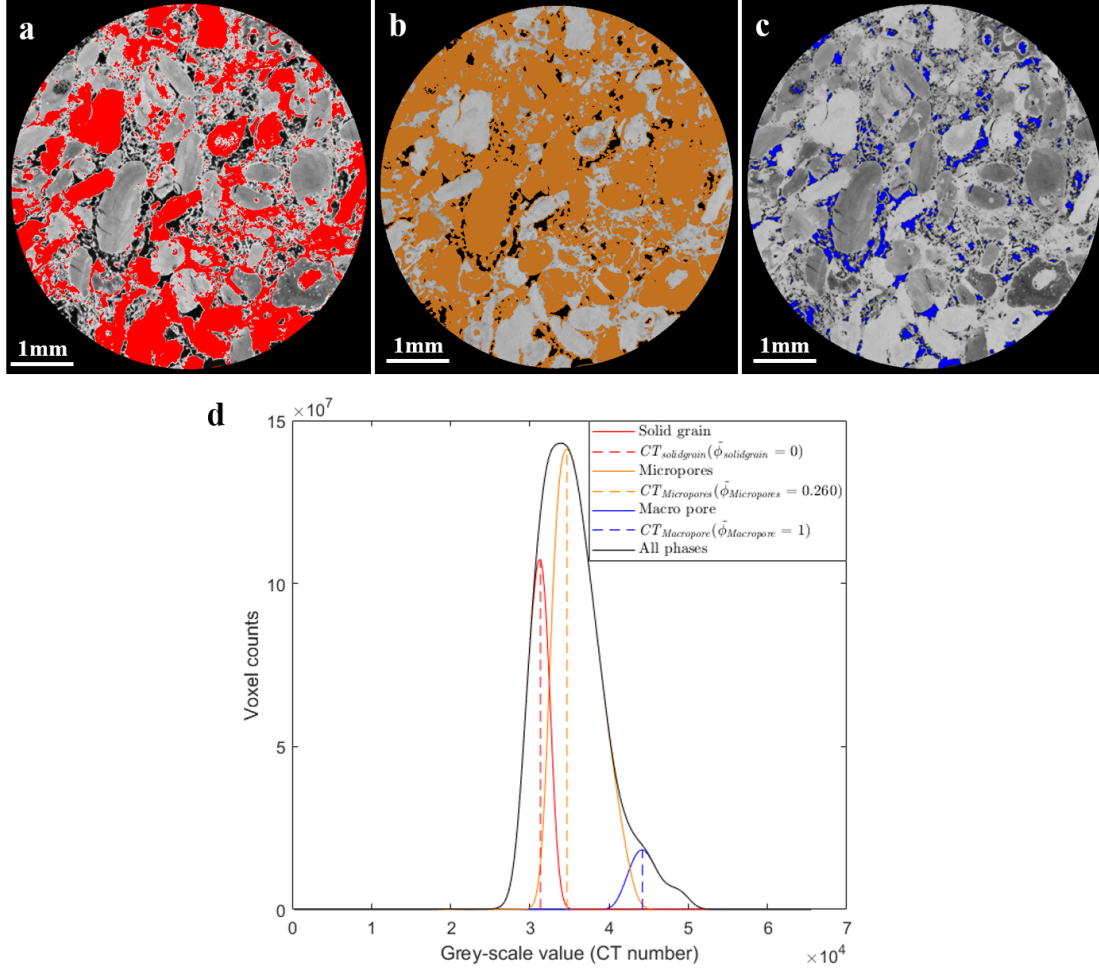


Figure 5.22: (a-c) Segmentation classification shown as two-dimensional cross-sections of the three-dimensional images of the same slice: (a) Solid grains (red). (b) Micro-pore phase (orange). (c) Macro-pore space (blue). (d) The histograms for the different phases in the dry scan. The red line represents the solid phase, the orange line represents the micro-porous phase and the blue line represents the brine phase. The dashed lines indicate the grey-scale values at the peak of each phase histogram.

where CT is the CT number of each voxel, and $CT0$ is a constant value to avoid negative grey-scale values. The difference images between the brine saturated scan and the multi-phase flow scan clearly identify changes in fluid occupancy. Figure 5.24 shows the difference images at different fractional flows for this experiment: black represents solid grain phase and unchanged brine, while white indicates oil in macro-pores. There are also grey regions which can represent either an intermittency, indicating that some of the time these regions were oil-filled and at other times brine-filled during the scan time of 40 minutes, or oil in the micro-porous phase, as shown in the red circles.

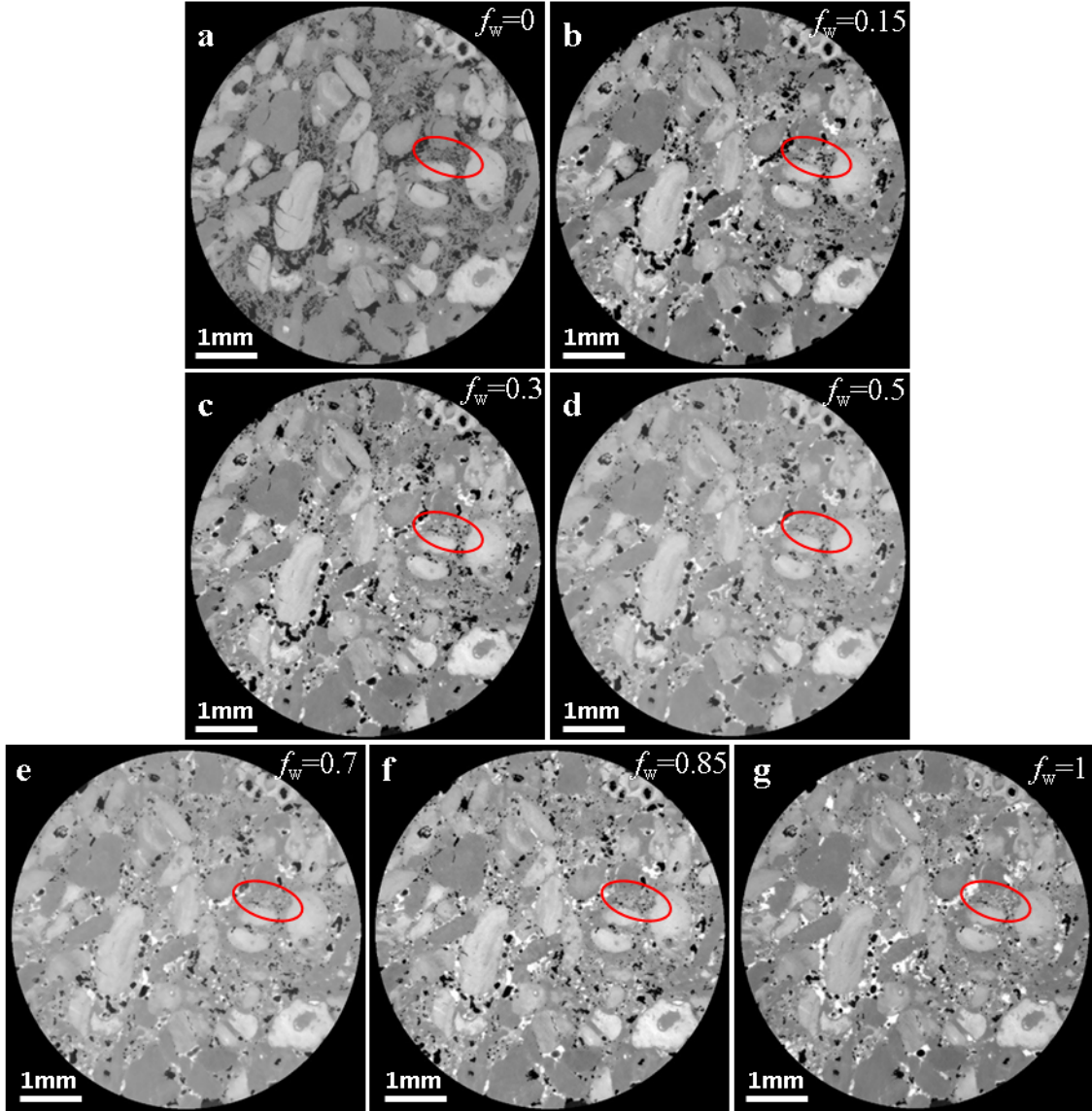


Figure 5.23: Two-dimensional cross-sections from three-dimensional micro-CT images of the same slice at different fractional flows. From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Grey includes grain, micro-porosity and intermittency in macro-pores, black represents stable oil in macro-pores which adsorbs X-rays the least, and white is brine in macro-pores which has the strongest X-ray adsorption. The red circles show areas where oil was displaced by brine in the micro-porosity.

5.3.3.1 Determination of fluid occupancy-based porosity

Based on the analysis of oil and brine pore occupancy at all fractional flows, we found that 85.5% of the micro-pore space was saturated only with brine, see Table 5.4. Oil did not occupy these pores because of the high capillary entry pressure. Hence, we distinguished four phases: solid grain, micro-pore phase I (always occupied by brine), micro-pore phase

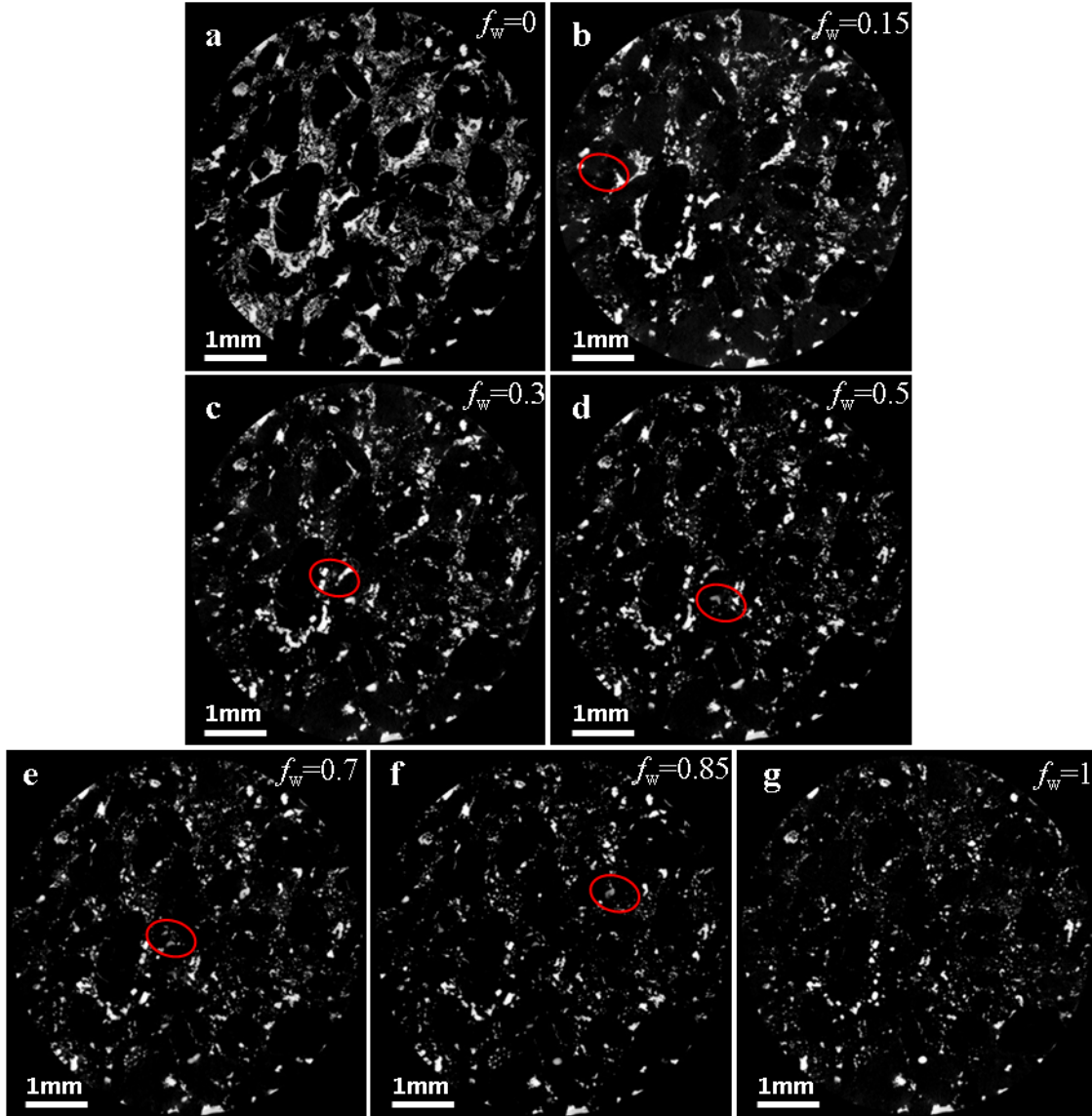


Figure 5.24: Two-dimensional cross-sections from three-dimensional difference images of the same slice. From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1. Black indicates solid grains and brine in the multiphase scans. White represents oil in the macro-pores. Grey represents either oil in micro-porosity, or an intermittent region (fluctuating pore occupancy of brine and oil) in the macro-pores.

II (which contains both oil and brine), and macro-pore space, as shown in Figure 5.25.

The histograms of CT number for all grey-scale voxels in the dry image are plotted for each phase, as shown in Figure 5.26. The red curve represents solid grain phase, green represents micro-pore phase I, the orange represents micro-pore phase II and blue represents macro-

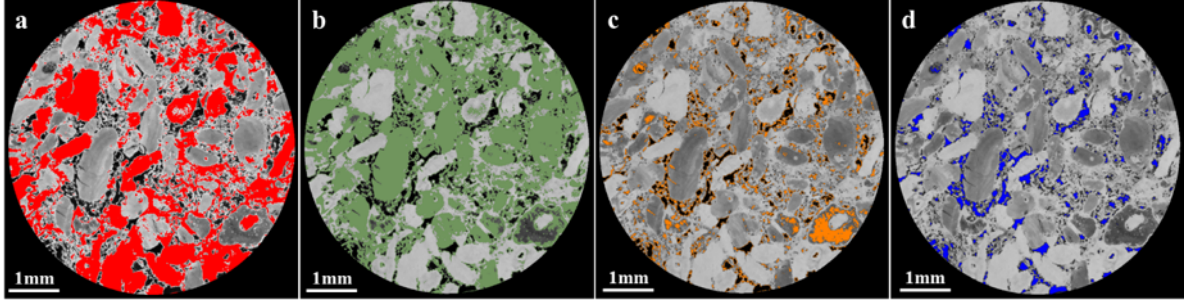


Figure 5.25: Segmentation classification shown on two-dimensional cross-sections of the three-dimensional images of the same slice. (a) Solid grain (red). (b) Micro-pore phase I (green), which is always saturated with water. (c) Micro-pore phase II, which contains both oil and water (orange). (d) Macro-pore space (blue), which can be saturated with either oil or water.

pore phase. In the dry image, the porosity of the micro-pore phase II is estimated as:

$$\phi_{microporeII} = p_{microporeII} \tilde{\phi}_{microporeII} = p_{microporeII} \frac{CT_{microporeII} - CT_{solidgrain}}{CT_{macropore} - CT_{solidgrain}} \quad (5.4)$$

where $\phi_{microporeII}$ is the porosity of micro-pore phase II, $p_{microporeII}$ is the fraction of the micro-pore phase II to the total pore volume, $\tilde{\phi}_{microporeII}$ is the porosity of the micro-pore phase II, and is calculated as 0.70 in this sample. The grey-scale values where the peak values are located for each pore space in the histogram are denoted as $CT_{solidgrain}$ for the solid grain phase, $CT_{microporeI}$ for the micro-pore phase I (occupied by brine only), $CT_{microporeII}$ for the micro-pore phase II (occupied by brine and/or oil), and $CT_{macropore}$ for the macro-pore phase.

The porosity of each phase measured using the differential imaging method is shown in Table 5.4.

Table 5.4: Volume fraction of solid grain, micro-pore space I, micro-pore space II and macro-pore space, and the corresponding porosity.

Type	Volume fraction p_i	Porosity $\tilde{\phi}_i$	Fraction of total volume of the sample ϕ_i
Solid grain	0.267	0	0
Micro-pore space I	0.568	0.260	0.148
Micro-pore space II	0.096	0.700	0.067
Macro-pore space	0.069	1	0.069
Total porosity	0.284		

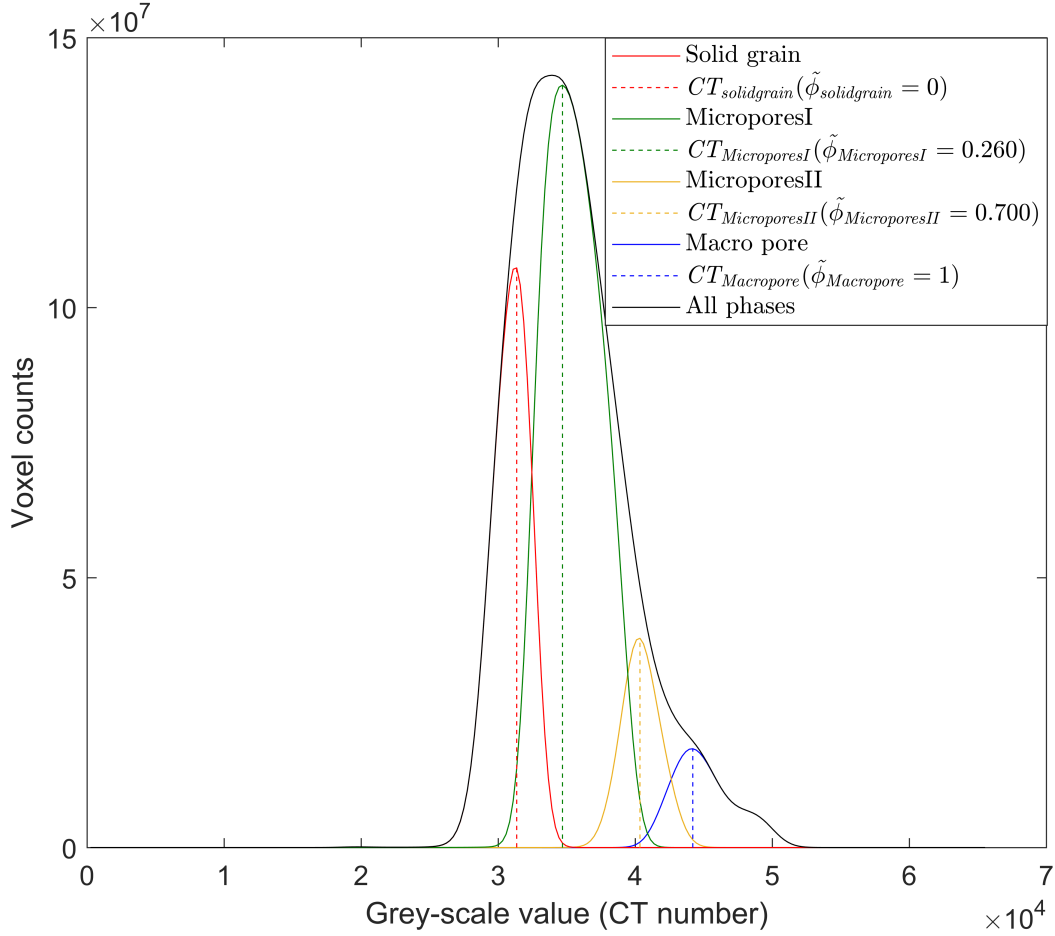


Figure 5.26: Histograms for the different phases of the dry scan. The red curve represents solid grain phase, green represents micro-pore phase I, orange represents micro-pore phase II and blue represents macro-pore phase. The red dashed line indicates the grey-scale value of the peak of the histogram for solid grain phase, the green dashed line indicates the grey-scale value of the peak of the histogram for the micro-pore phase I, the orange dashed line indicates the grey-scale value of the peak of the histogram for the micro-pore phase II, and the blue dashed line shows the grey-scale value of the peak of the histogram for the macro-pore phase.

5.3.3.2 Fluid occupancy in micro-pore space

Flow heterogeneity in carbonates is affected by the presence of micro-porosity. Using the differential imaging method, we distinguish pore occupancy in both the macroscopic and microscopic pore space. The raw images were segmented based on the four-phase masks shown in Figure 5.25 and the difference images shown in Figure 5.24. Furthermore, oil, brine and partially filled micro-pores were segmented based on the grey-scale distribution histogram of micro-pore space II, shown in Figure 5.27.

In order to minimize the uncertainties due to counting statistics and sample position variations, all the images have been assigned and resampled at the same orientation and same size. This shows that sample position variation is not a problem. Also, the grey scale value distribution of the images was normalised to ensure that the peak values of brine and grain phase at different images were the same. This helps to decrease the uncertainty due to counting statistics. As the grey scale value is higher for water than the value for grains which in turn is larger than the value for oil, the grey scale of water in micro-pores is larger than that of oil in the micro-pores. This method assumes that water saturation in the sub-resolution porous space is characterised by a distribution of grey-scale values representing water and oil saturation values.

For this method, the main uncertainties can be caused by two factors. 1) The classification of micro-pore space, shown in Figure 5.25. The pore size distribution of the micro-pore phase can influence the results. As the pore size of Estailades carbonate has a wide distribution, we classified the micro-pores into two groups: micro-pore phase I and micro-pore phase II. For more complex rocks, the micro-porosity could be broken down into more than two regions. 2) The image quality also affects the results. Theoretically, the image quality can be increased by increasing scan time. However, this time cannot be too long because rearrangement of fluids in pore space needs to be captured. Thus we balanced the image quality and scan time in this work.

The positive point is that this is a consistent method to measure the oil/water saturation in micro-pores based on micro-CT images. The limitations of this work are that: 1) I only divide the micro-porous space into two sub-phases. If more sub-phases were divided, the saturation may be computed more accurately, especially for a rock with a wide pore size distribution; 2) the image quality is not perfect because the scan time has to be considered. If the flow of this experiment was at low capillary number, a longer scan can be taken.

The results provide the likely uncertainty in our measured saturations and pressure differences, taking into account different possible ways to threshold the micro-porous and intermittent regions, which are captured in the error bars shown when we plot relative permeability.

In the multiphase images the contribution to the brine saturation in the partially filled

micro-pores was estimated as:

$$S_{w,PF,microporeII} = \phi_{PFM} \tilde{S}_w = \phi_{PFM} \frac{CT_{PFmicropores} - CT_{oil}}{CT_{brine} - CT_{oil}} \quad (5.5)$$

where $S_{w,PF,microporeII}$ is the contribution to the brine saturation in partially filled micro-pores in micro-pore space II, ϕ_{PFM} is the fraction of the partially filled micro-pore space to the total pore volume of micro-pore space II. The grey-scale values where the peak values are located for each phase in the histogram are denoted as CT_{oil} for the oil in micro-pore space II, $CT_{PFmicropores}$ for the partially filled micro-pores, and CT_{brine} for the brine in micro-pore space II, and $\tilde{S}_w$ denotes the proportion of brine in partially filled micro-pores in micro-pore space II.

For a fractional flow of 0.5, the fraction of brine in partially filled micro-pore space is 0.333 (shown in Figure 5.27) and the fraction of the partially filled micro-pores in the total pore space in micro-pore space II is 0.192 (shown in Table 5.5), so the contribution to the total brine saturation from partially filled micro-pores in micro-pore space II is the product of these two numbers giving the value of 0.064. Fluid occupancy in micro-pore space for all fractional flows is provided in Table 5.5.

5.3.4 Fluid occupancy in the macro-pore space

The difference images, shown in Figure 5.24, were segmented to obtain the oil phase (white), and the intermittency (grey) in the macro-pores. Then the brine phase was obtained by subtracting the oil and intermittent phase from the macro-pore phase. These three phases in macro-pores, shown in Figure 5.28, were segmented into brine (blue), oil (red) and intermittent (yellow) phases.

Using these segmented label images as reference, separate raw images for each phase were extracted. Figure 5.29 shows the histograms of the grey-scale values of the image at a fractional flow of 0.5, for the oil, brine and intermittency of the macro-pore space. In the macro-pore space, the contribution of the intermittent phase in the macro pores to the water saturation was estimated as:

$$S_{w,inter,macropore} = \phi_{inter} \tilde{S}_w = \phi_{inter} \frac{CT_{intermittency} - CT_{oil}}{CT_{brine} - CT_{oil}} \quad (5.6)$$

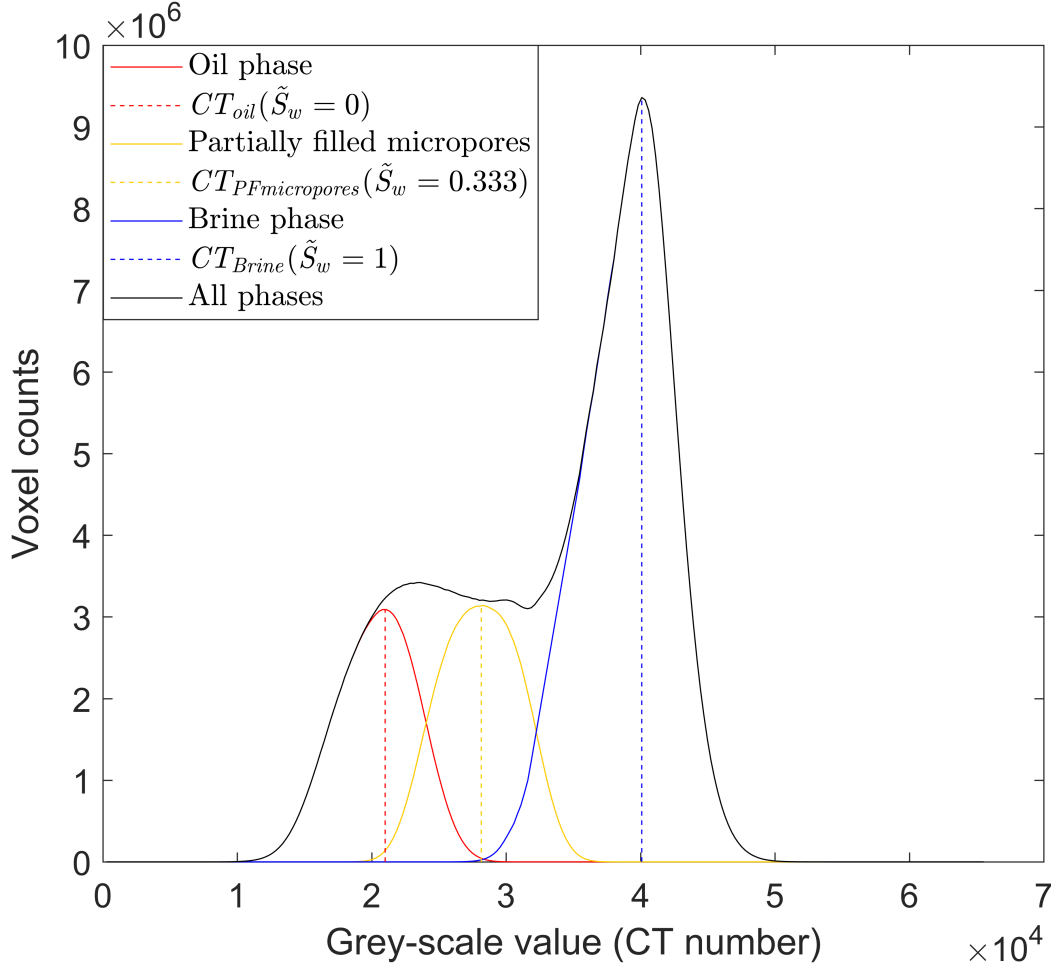


Figure 5.27: Histograms of the grey-scale values for the oil, partially filled micro-pore and brine phase in micro-pore space II when $f_w = 0.5$. The dashed lines in the histogram indicate the grey-scale values where the peaks for each phase exist. For the partially filled micro-pores the proportion of brine is 0.333 (calculated using Equation (5.5)), while the contribution to the total brine saturation in partially filled micro-pores in micro-pore space II is 0.064.

where $S_{w,inter,macropore}$ is the contribution of the intermittency to the brine saturation in macro pores, and ϕ_{inter} is the fraction of the intermittency to the total pore volume in macro pores. The grey-scale values where the peak values are located for each phase in the histogram were denoted as CT_{oil} for the stable oil phase, $CT_{Intermittency}$ for the intermittency, CT_{brine} for the stable brine phase, and $\tilde{S}_w$ denotes the proportion of brine in the intermittency.

For illustration, for a fractional flow of 0.5, the fraction of brine in the intermittency is 0.470 (shown in Figure 5.29) and the fraction of the intermittency in macro pore space is 0.237 (shown in Table 5.5), hence the contribution to the total brine saturation in the

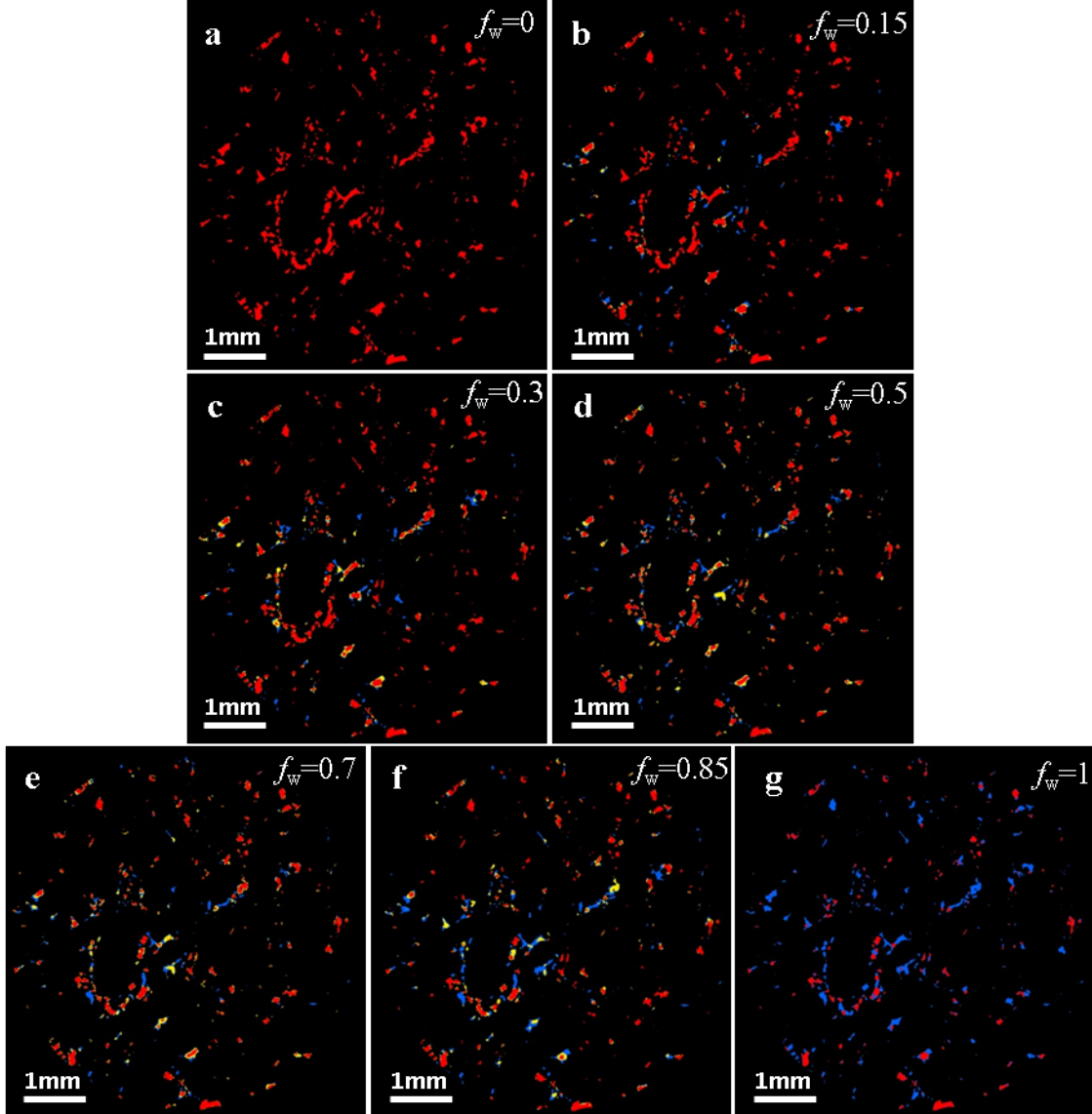


Figure 5.28: Three-phase segmented label image showing brine (blue), intermittent phase (yellow) and oil phase (red) in the macro-pore space. From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1.

intermittency in macro-pore space is 0.111. In the next section we provide fluid occupancy in the macro-pore space for all fractional flows.

5.3.5 Fluid occupancy in the macro- and micro-pore space and dependence on fractional flow

The brine saturation in the micro-pore space, macro-pore space, and the total saturation along the image are shown in Figure 5.30. The uncertainty in saturation averaged over

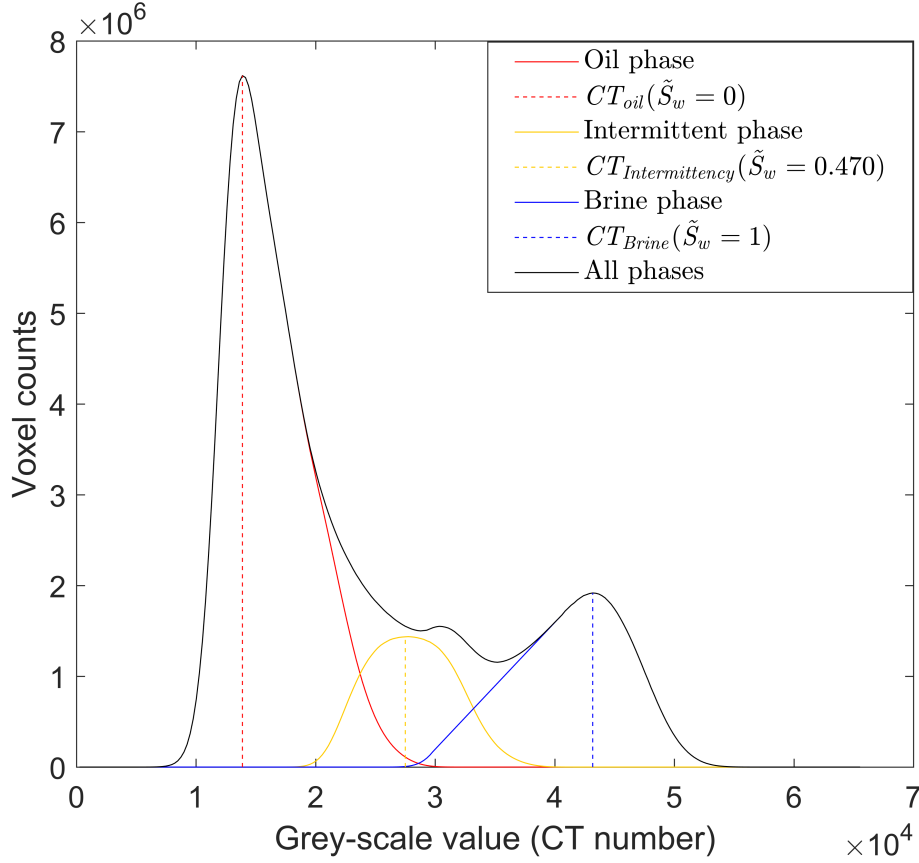


Figure 5.29: The histograms for the different phases in macro-pore space when f_w is 0.5. The dashed lines in the histograms indicate the grey-scale values where the peaks for each phase exist. The proportion of brine in the intermittency is 0.470 (calculated using equation (5.6)), while the contribution to the total brine saturation in the intermittency in macro-pore space is 0.111.

each slice for total pore space is approximately 3%, including 2% for micro-pores and 1% for macro-pores. The amount of brine in the macro-pore space was approximately constant. However, the fraction of brine in micro-pore space varied along the image because Estailades has a heterogeneous pore structure. Overall, the brine saturation was not uniform along the image because micro-porosity made a significant contribution to the total brine saturation.

From the profiles we observe that a considerable fraction of micro-porosity became occupied by brine that displaced oil when the fractional flow was initially increased from 0 to 0.15. Further increases resulted in a smaller oil displacement that was non-uniform along the core, reflecting the complex micro-porous space. These findings illustrate that in a water-wet rock, pore filling during a waterflood occurs in order of increasing size, and hence the micro-porosity is preferentially filled first. This is further confirmed from the plots of

Table 5.5: Percentages of the stable oil, stable water and intermittency, as well as the water saturation. Also shown is the fraction of the oil that connects from inlet to outlet, hence allowing flow and displacement, considering just the stable oil phase, and for the stable oil and intermittent phase combined.

f_w	$Ca = 7.3 \times 10^{-6}$							
	S_w	Fraction of the macro-pore space containing stable oil	Fraction of the macro-pore space containing stable water	Fraction of the macro-pore space containing intermittency	Fraction of micro-pore phase II that is partially filled	Fraction of micro-pore phase II filled with oil	Fraction of micro-pore phase II filled with water	The water saturation S_w from micro-pore phase I
0	0.588	0.988	0.012	0.000	0.000	0.723	0.277	0.521
0.15	0.702	0.753	0.115	0.132	0.170	0.307	0.523	0.521
0.3	0.720	0.666	0.128	0.206	0.142	0.299	0.559	0.521
0.5	0.765	0.578	0.185	0.237	0.192	0.191	0.617	0.521
0.7	0.779	0.564	0.181	0.255	0.194	0.186	0.620	0.521
0.85	0.781	0.513	0.276	0.211	0.164	0.177	0.659	0.521
1	0.855	0.362	0.638	0.000	0.000	0.240	0.760	0.521

remaining oil saturation in micro- and macro pore space shown in Figure 5.31. The oil saturation decreased faster in the micro-porous regions with smaller pores until a fractional flow of 0.85.

Table 5.5 lists the fractions of oil and water in the macro- and micro-porous regions. As the fractional flow increases, water displaces oil.

5.3.6 Relative permeability

As mentioned in Section 4.1.1.2, the pressure difference across the rock sample at steady-state conditions was measured using the differential pressure transducer shown in Figure 4.4. Figure 5.32 shows the mean pressure differentials at different fractional flows during the last two hours of the total 15 - 24 hours, as well as the corresponding standard deviation. We have subtracted the pressure drop in the flow lines themselves.

When the fractional flow was 0 and 1, the pressure profile was smooth, which indicates an unchanging fluid configuration. However, there were rapid pressure fluctuations during intermediate fractional flows, as observed previously (Sinha and Hansen, 2012; Sinha et al., 2017). For this experiment, the range of the fluctuations was from 0.55 kPa to 8.63 kPa. In the Bentheimer experiment at the same Ca (Gao et al., 2017), see Section 5.2, the range was from 0.12 kPa to 10.72 kPa. The pressure fluctuations are indicative of pore-

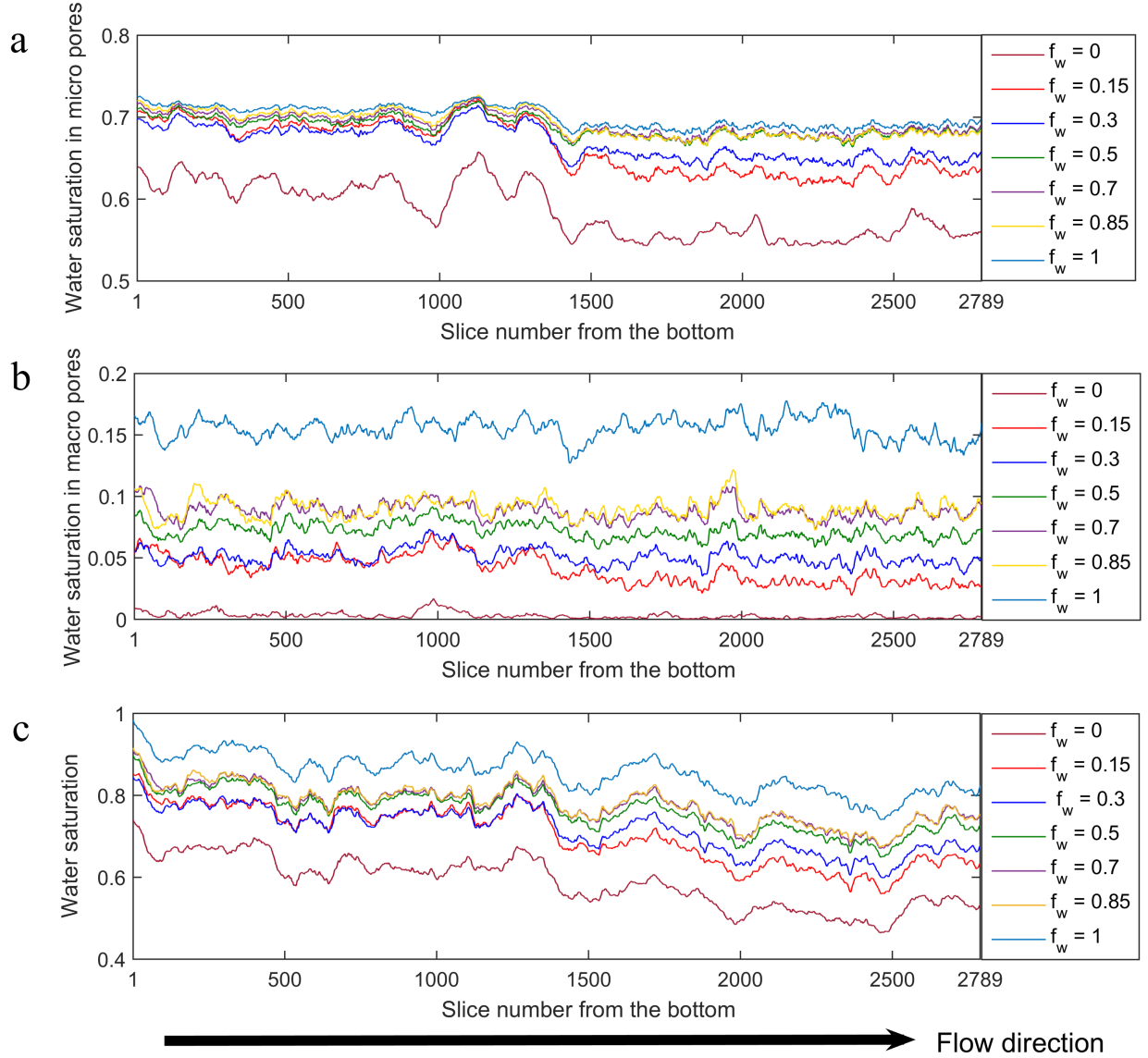


Figure 5.30: Brine saturation profiles in micro-pores (a), macro-pores (b) and total saturation (c). From bottom to top on each graph, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1.

scale rearrangements and have a magnitude equal to a typical capillary pressure, which is approximately 12.5 kPa calculated from $2\sigma/r$ where σ is the interfacial tension and r is the average throat radius of $7.5 \mu\text{m}$ for Estailades (Blunt, 2017).

The absolute permeability measured using the same transducer was $1.06(\pm 0.02) \times 10^{-13} \text{ m}^2$. The relative permeability was calculated using (as for Bentheimer, section 5.2):

$$k_{rw} = \frac{Q_w \mu_w L}{A \Delta P k}; k_{ro} = \frac{Q_o \mu_o L}{A \Delta P k} \quad (5.7)$$

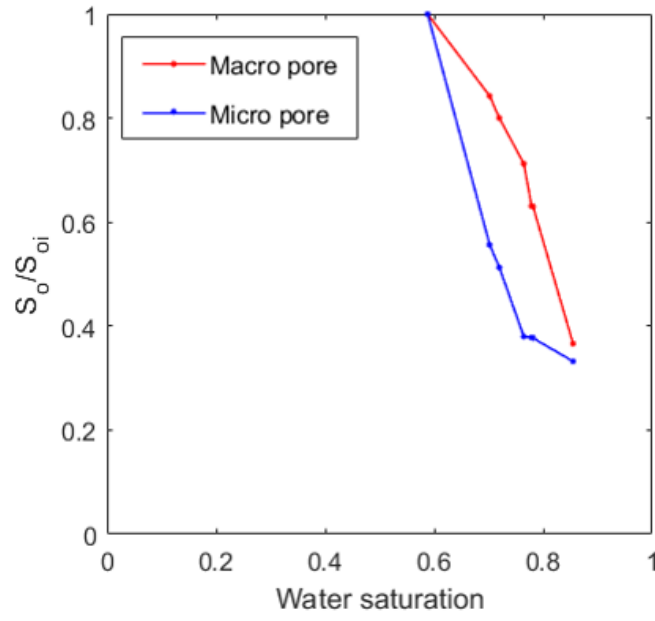


Figure 5.31: Oil saturation normalised by initial oil saturation in the micro- and macro-pore space as a function of total water (brine) saturation.

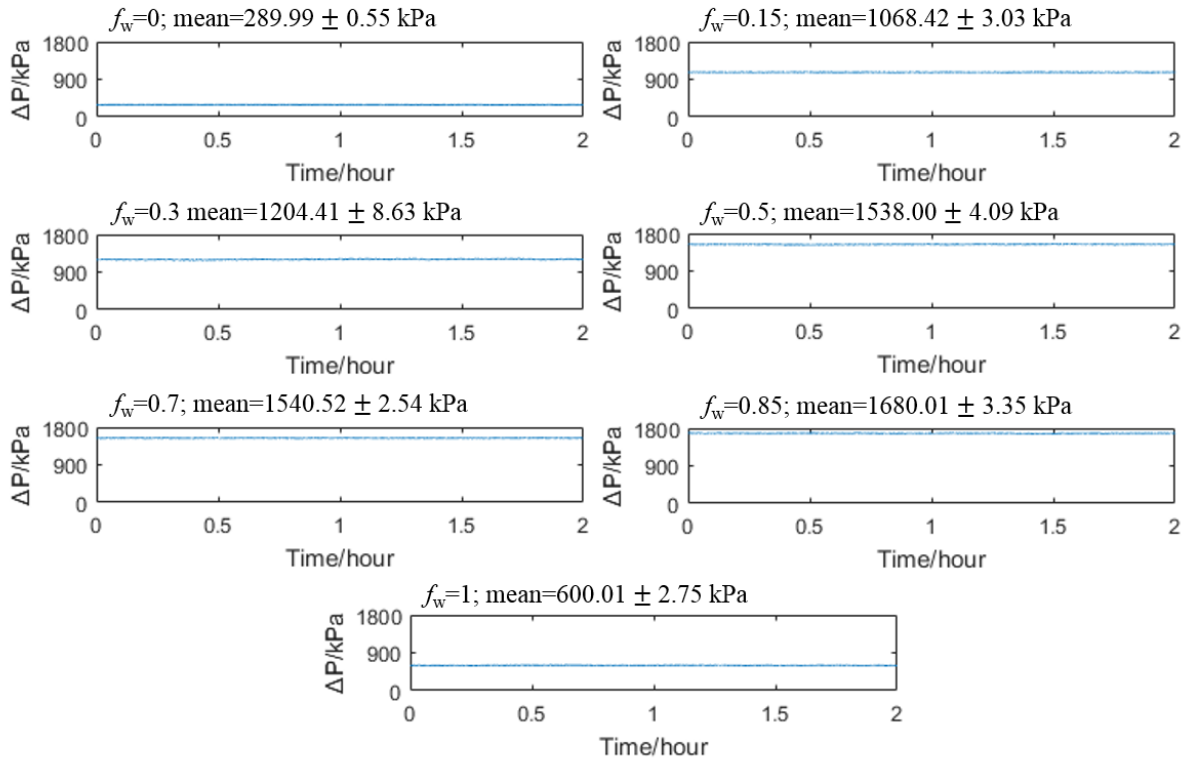


Figure 5.32: Pressure difference measured between the inlet and outlet of the samples. The mean pressure drop and standard deviation at each fractional flow are shown.

where k_{rw} is brine relative permeability, k_{ro} is oil relative permeability, Q_w is brine flow rate, m^3/s , Q_o is oil flow rate, m^3/s , μ_w is brine viscosity, $\text{Pa}\cdot\text{s}$, μ_o is oil viscosity, $\text{Pa}\cdot\text{s}$, L is the length of the whole sample, m , A is the cross area of the sample, m^2 , k is absolute permeability, m^2 , and ΔP is the pressure difference across the sample, Pa . We do not need to correct for capillary pressure gradients, as there is no discernable saturation gradient across the core, particularly considering the macro-pore space, see Figure 5.30 (Lin et al., 2018).

Table 5.6 shows the wetting phase saturation and the measured oil and brine relative permeabilities at each fractional flow. The saturation values shown in the table were calculated from the scanned region of the core.

Table 5.6: Saturation and relative permeability calculated over a range of f_w .

f_w	$Ca = 7.3 \times 10^{-6}$			
	S_w	k_{ro}	k_{rw}	$\Delta p / \text{kPa}$
0	0.588	0.577	0.000	289.99 ± 0.55
0.15	0.702	0.133	0.023	1068.42 ± 3.03
0.3	0.720	0.097	0.041	1204.41 ± 8.63
0.5	0.765	0.053	0.053	1538.00 ± 4.09
0.7	0.779	0.032	0.075	1540.52 ± 2.54
0.85	0.781	0.019	0.083	1680.01 ± 3.35
1	0.855	0.000	0.274	600.01 ± 2.75

Figure 5.33 shows the estimated fractional flow curves at steady state for Estailades carbonate studied here and Bentheimer sandstone shown in Section 5.2 (Gao et al., 2017) together with the relative permeabilities for these two rocks. The uncertainties in saturation were estimated from the root-mean square uncertainties in phase volume, total volume and porosity during image segmentation in both figures. The uncertainties in relative permeability consider uncertainties in the measurement of the length and diameter of the sample, the absolute permeability, pressure measurements, accuracy of pressure transducers and the pump rates. At the end of primary drainage, the initial brine saturation for Estailades was 0.588 ± 0.020 , while at the end of imbibition, the residual oil saturation was 0.145 ± 0.016 . The initial brine saturation for Bentheimer was 0.145 ± 0.031 , while the residual oil saturation was 0.337 ± 0.030 .

Both the sandstone and carbonate samples are strongly water-wet. The micro-porosity has a significant impact of on the relative permeability of Estailades carbonate. Since brine

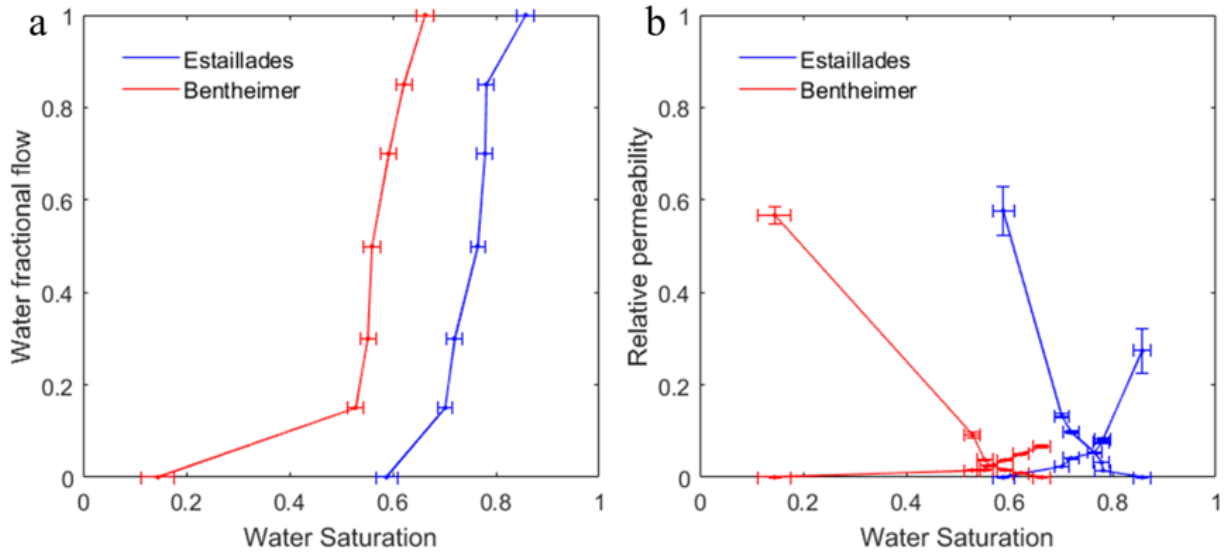


Figure 5.33: (a) Fractional flow curves calculated at steady state for water (brine)-oil imbibition for Estailades (blue) and Bentheimer (red) from Gao et al. (2017). (b) Relative permeability plotted as a function of wetting phase (water/brine) saturation in Estailades and Bentheimer (Gao et al., 2017). Error bars in the x-direction consider uncertainties in the measured saturation caused by the threshold value chosen during image segmentation. Error bars in the y-direction consider uncertainties in the measurement of the length and diameter of the sample, the absolute permeability, pressure measurements, accuracy of pressure transducers and the pump rates.

occupies micro-pores there is a considerable shift to higher brine saturations compared to Bentheimer sandstone. Moreover, the Estailades water (brine) relative permeability rises more steeply and to higher values, which is an indication of the enhanced permeability and connectivity from micro-porous regions. The residual oil saturation is lower than in the sandstone; this is partly due to the higher initial brine saturation and also due to additional connectivity through oil-filled micro-porosity preventing complete disconnection of the oil. In contrast, the oil, that only occupies a small fraction of the micro-porosity, has a relative permeability that is similar in shape to the sandstone, albeit shifted to higher brine saturation.

5.3.7 Pore and throat occupancy of the intermittency

We now test the hypothesis that the existence of the intermittent flow in the macroscopic pore space depends on pore and/or throat size. A generalized pore network extraction algorithm (Raeini et al., 2017) was used to subdivide macroscopic pore space into pores

bounded by throat surfaces. Each voxel was assigned to a unique pore as well as to a unique throat (the closest throat surface). The volume of the pore and throat voxels, the inscribed radii were used to analyse the statistics of the pore and throat fluid occupancy.

The pores and throats in the network were sorted based on their radii and divided into bins of equal volume. These bins, whose centres are filled with oil, water or the intermittency, were then used to compute the volume fraction of pores and throats. Figure 5.34 shows the results for different fractional flow images. As expected for a water-wet medium, the oil

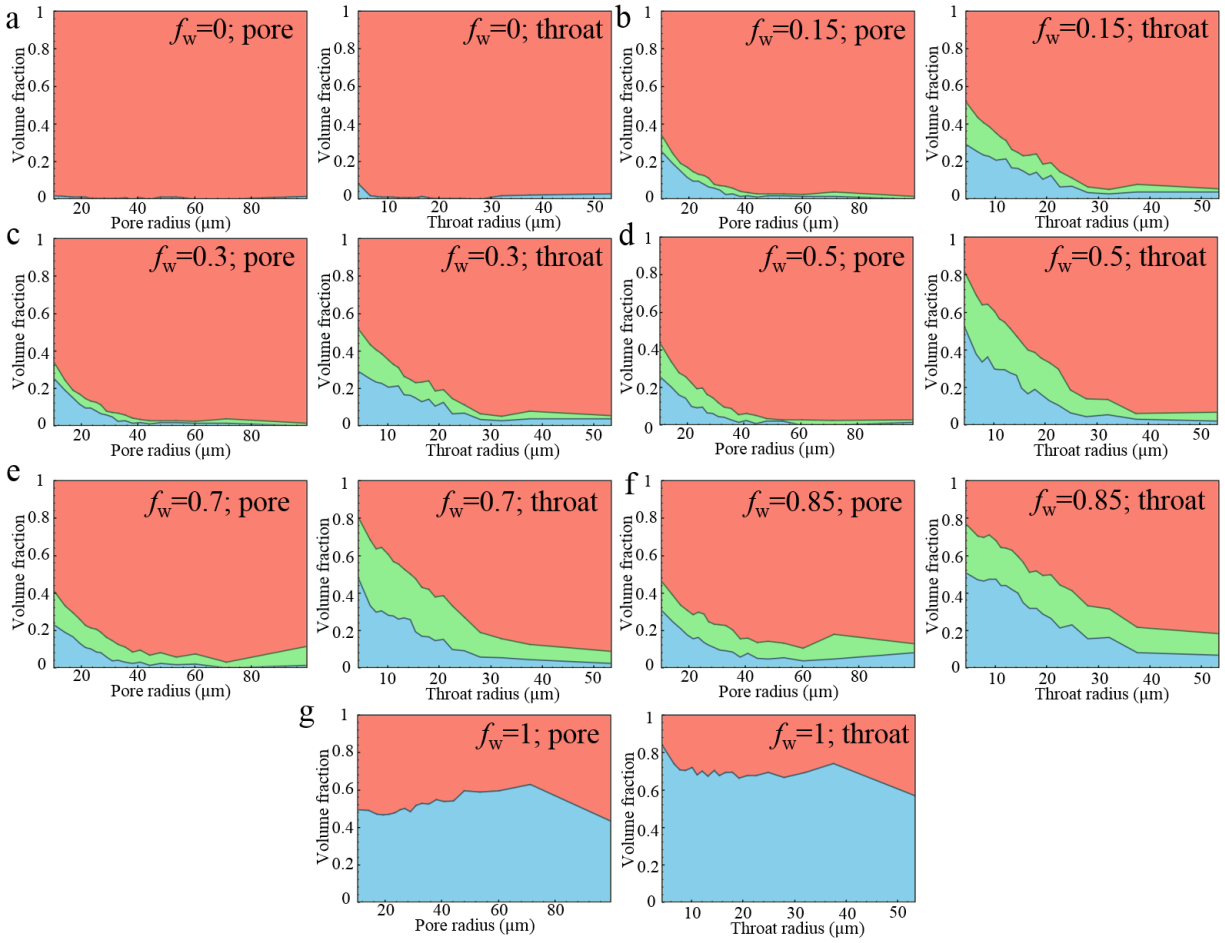


Figure 5.34: The volume fraction of radii of pores and throats occupied by oil, water and intermittent phases as a function of radius. Red represents oil, green is the intermittency, and blue is brine.

occupies the larger pores while brine preferentially resides in small pores and throats. There is more water in throat centres than in pore centres, as throats represent restrictions in the pore space. However, water is seen in some larger elements, and in general the tendency for water to reside strictly in the smaller pores and throats is less marked than that observed

in water-wet sandstones (Bultreys et al., 2018). In particular, when the oil is trapped (for $f_w = 1$) we see oil and water in elements of all sizes with only a weak preference for oil to remain in larger pores. The reason for this is likely to be the presence of micro-porosity. As shown in Table 5.5, some oil also resides in the micro-porosity. Hence a much wider range of pore sizes are occupied by both phases than encountered typically in sandstones: the exact occupancy will be controlled by pore size, local pore geometry and connectivity, and we even see some oil trapped in the micro-porosity at the end of waterflooding.

As evident in Figure 5.34, we mostly see the intermittent flow for intermediate fractional flows: for $f_w = 0$ and $f_w = 1$, the flow paths are fixed, as only one phase is flowing. However, when both phases simultaneously flow through the rock, there are regions of the pore space where – during the period of the scan – sometimes oil is present and sometimes brine. This shows that the flow pathways are not fixed, but that a dynamic or intermittent connectivity is observed. This fluctuating occupancy has been seen in Bentheimer sandstone in Section 5.2 and observed directly using fast synchrotron imaging (Reynolds et al., 2017). Compared to the sandstone, however, the fraction of the pores occupied by the intermittency in the carbonate was smaller. For example, when $f_w = 0.5$, the fraction of the pores occupied by the intermittency in the macro-pore space was 0.24 in Estailades, while it was 0.33 in Bentheimer sandstone. The intermittent flow is seen mainly in the intermediate-sized and smaller macro-pores, as opposed to only the intermediate sized pores in a sandstone.

The physical explanation of intermittency is that the flowing fluids have sufficient energy to periodically open more conductive flow channels (Reynolds et al., 2017): at lower flow rates, there is insufficient energy to overcome the capillary pressure barriers that block short-cuts in the flow. In a carbonate, oil and water are also present in micro-porosity, and so it is not surprising that even some of the smaller macro-pores allow intermittent connectivity at moderately high flow rates, facilitating flow through both the macro- and micro-porous regions.

5.3.8 Oil ganglia distribution analysis

Percolation theory assumes that the pore filling by water is in order of size, resulting in a prediction of the trapped ganglia size distribution. According to percolation theory, the number n of clusters of volume s (measured in voxels) should scale as $n(s) \sim s^{-\tau}$, where τ

is the Fisher exponent (Fisher, 1967). Andrew (2014) performed unsteady-state waterflood experiments displacing supercritical CO₂ for a range of sandstones and carbonates. They obtained $\tau = 2.28$ for the residual CO₂ trapped in the pore space of Estailades carbonate. However, as shown in Figure 5.35, trapping is not strictly in the larger macro-pores and so it is not evident that the percolation predictions hold in this case.

Here, we analyse the ganglion size distributions for each fractional flow, imaged after reaching steady state in the experiment. We ignored ganglia smaller than 10^2 voxels, which is much less than the size of a single pore. We only considered the macro-pore space and also ignored the intermittency. Following Dias and Payatakes (1986), we define:

$$N_s = \sum_{s'=s}^{2s-1} n_{s'} \quad (5.8)$$

which should scale as:

$$N_s \sim s^{-(\tau-1)} \quad (5.9)$$

N_s as a function of s is plotted on logarithmic axes in Figure 5.35, showing approximate power-law scaling behaviour for ganglia larger than 10^4 voxels. The best-fit exponents were then calculated by performing a Levenberg–Marquardt regression (Levenberg, 1944; Marquardt, 1963) using a least absolute residual robust fitting algorithm (Holland and Welsch, 1977; Huber, 2011). Figure 5.36 shows three-dimensional visualizations of the ganglia which are divided into different categories based on their size: large ganglia, which typically contain tens to hundreds of pores and may be connected across the field of view (red volumes, containing more than 10^5 voxels); medium ganglia, which may be connected over tens of pores (blue volumes, containing 10^4 to 10^5 voxels); and small ganglia, which may be isolated in a single pore or span over a few pores (yellow volumes, containing below 10^3 to 10^4 voxels); ganglia smaller than 10^3 are not shown.

Table 5.7 shows the fitted Fisher exponents at different fractional flows. The calculated exponent is in the range between 1.92 and 2.24. When the fractional flow is 0, the Fisher exponent is the smallest which is due to the high connectivity of the non-wetting phase. At higher fractional flows, the ratio of small ganglia to large ganglia is higher, which can also be observed in Figure 5.36, as more small (yellow) and intermediate (blue) but less large (red) ganglia occupy the pore space. Strictly, percolation theory is only valid for fully trapped phases, when $f_w = 1$ and all the oil is trapped: in this case the value obtained for

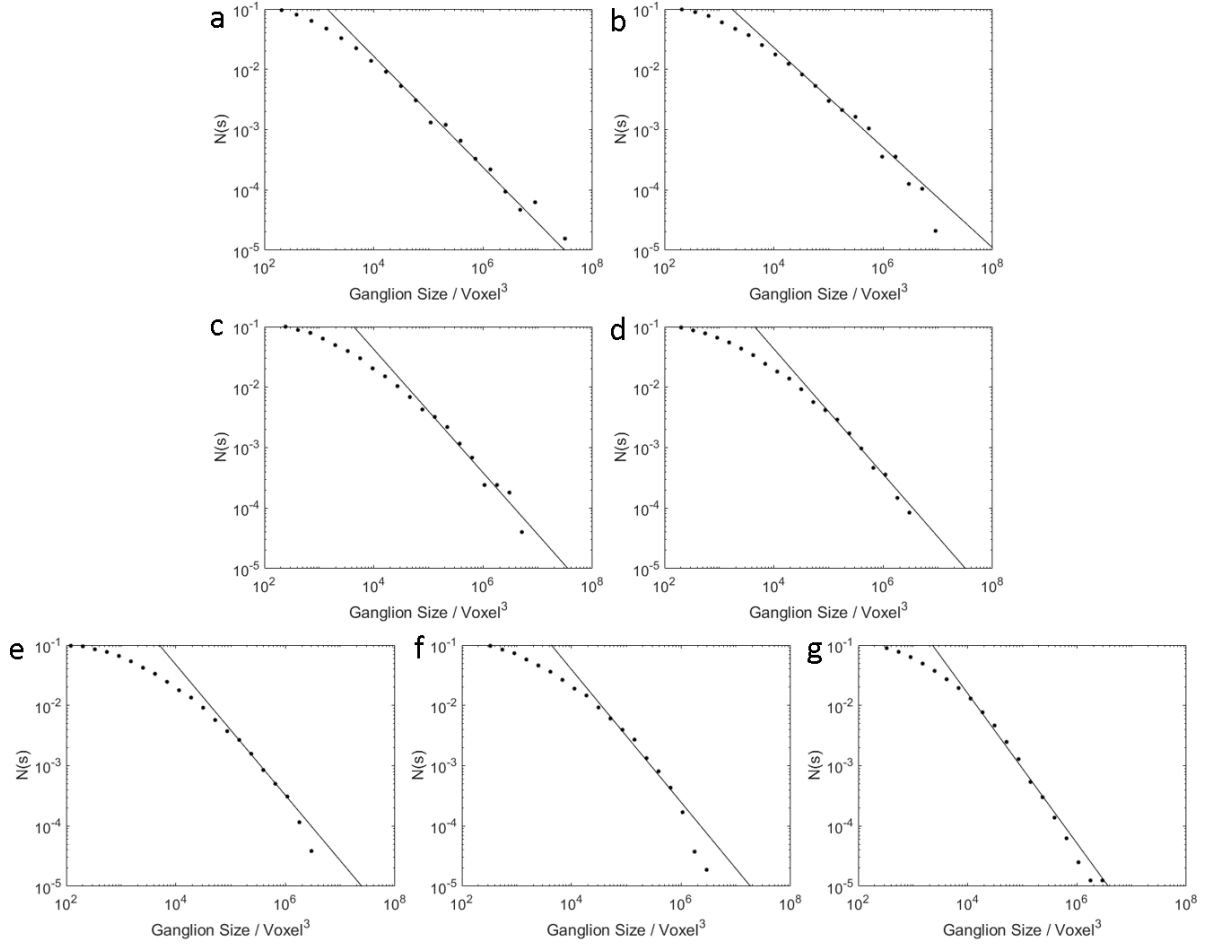


Figure 5.35: Ganglia size distribution, N_s . From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1.

the exponent is consistent with previous studies on Estailades for CO_2 ganglia trapped by brine (Andrew et al., 2014).

Table 5.7: Fisher exponents for the non-wetting phase ganglia sizes at different fractional flows.

f_w	0	0.15	0.30	0.50	0.70	0.85	1
τ	1.92 ± 0.04	1.93 ± 0.02	2.02 ± 0.05	2.04 ± 0.01	2.08 ± 0.01	2.10 ± 0.04	2.24 ± 0.04

5.3.9 Summary

We have established a novel experimental and image analysis methodology to measure the pore occupancy, relative permeability and flow intermittency in a complex micro-porous carbonate. We have demonstrated this by conducting steady-state waterflood experiments

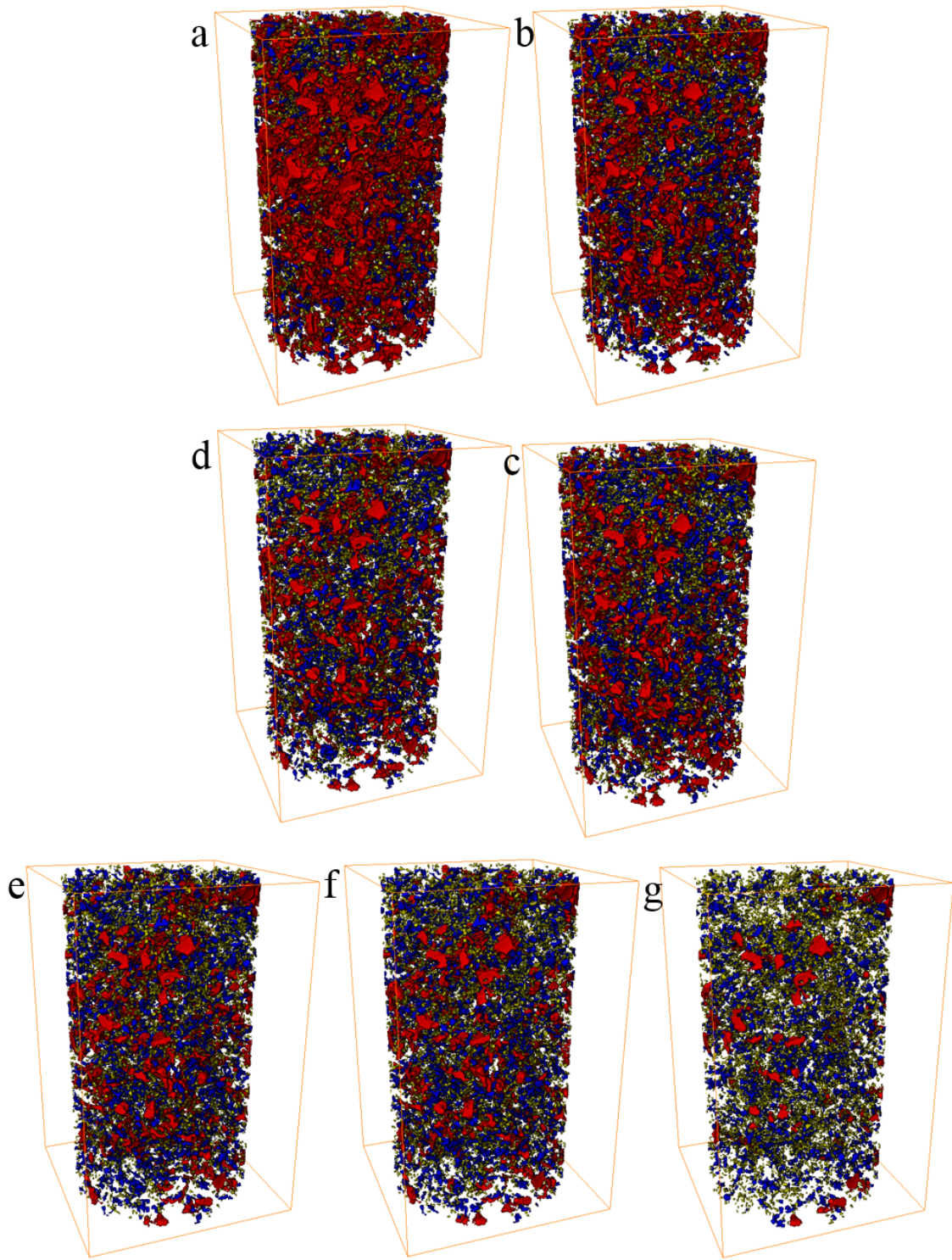


Figure 5.36: Non-wetting phase ganglia populations contributing at each fractional flow. Ganglia are subdivided by size: large (red), medium (blue), and small (yellow). Flow is from bottom to top. From left to right and from top to bottom, the fractional flow is 0, 0.15, 0.3, 0.5, 0.7, 0.85, and 1.

on an Estailades limestone sample at a capillary number, $Ca = 7.3 \times 10^{-6}$, and performing differential imaging to distinguish macro-pore space, explicitly resolvable in the image, and micro-porosity, together with fluid occupancy.

Estailades is a heterogeneous carbonate composed of approximately 27% solid grains, 7% macro-pores, and 66% micro-pores that could not be explicitly resolved from micro-CT images. We were able to quantitatively describe four phases: solid grain, pores occupied only by brine (micro-pore space I), micro-pores partially filled by oil and brine (micro-pore space II), and macro-pore space, which itself can contain oil, water or can be intermittent. During multiphase flow we could distinguish three types of fluid occupancy in the micro-pore space II: filled with oil, filled with brine, and partially-saturated with both brine and oil present. The micro-porosity contributed significantly to the total porosity: the fully brine-saturated portion of the pore space, approximately 57% of the total pore volume of the rock, contributed to a large initial brine saturation after primary drainage, shifting the relative permeabilities to the right compared to Bentheimer sandstone. However, some of the micro-porosity, approximately 10% of the whole sample, initially contained oil that was displaced during waterflooding: the enhanced connectivity of the pore space through micro-porosity led to a higher end-point water relative permeability and a lower residual oil saturation than observed for a sandstone under similar conditions. Our method indicated initially higher recovery from micro-porosity which consists of smaller pores. This is expected in a water-wet rock where the wetting phase preferentially fills the smaller pores.

In the macro-pore space, the intermittent flow was identified by the differential imaging method, showing that there were some regions of the pore space that were occupied alternately by oil and brine. The intermittent region was seen principally in the smaller and intermediate sized pores and the fraction of the pore space showing this fluctuating occupancy was slightly smaller than in Bentheimer sandstone under the same conditions.

This experimental technique is readily applicable to other systems in which micro-porosity plays an important role, including mixed-wet carbonates. Future work could include a wide variety of the pore-scale multiphase flow problems, including those at the conditions representative of subsurface aquifers, oil and gas fields and other deep geological systems.

In next chapter, dynamic flow regimes are explored with the state-of-the-art synchrotron imaging facility to observe displacement processes directly.

Chapter 6

Dynamic multiphase fluid flow imaged using synchrotron X-ray micro-tomography

The work presented in this chapter is a pore-scale experimental investigation of microscopic steady-state flow during co-injection from very low to high flow rates in the pore space of a sandstone using 4D synchrotron X-ray micro-tomography to advance our understanding of flow regimes. We investigate rate dependent, steady-state two-phase flow behaviour at a series of capillary numbers representative of subsurface flow conditions, covering the capillary numbers below and above the threshold for pore-scale ganglion motion. The measured pressure differences across the sample are measured at different flow rates. This is a natural extension of the work of Reynolds et al. (2017), in which we study an oil/brine system, rather than gas/brine, and explore a wider range of flow rates coupled with pressure measurements.

The purpose of this experiment was to investigate the effect of viscous forces on the onset of intermittent effects in multiphase flow. As described in Chapter 5, we have already seen that intermittency occurs when Ca is approximately 7×10^{-6} using laboratory-based micro-CT but now we wish to study fluid behaviours for a range of capillary numbers to elucidate the dynamics and quantify the range of Ca when these effects become important.

In this work, to help place the work in context, and before presenting a detailed analysis of the imaging results themselves, we hypothesise possible flow regimes in Section 6.1. Then the experiment is described in Section 6.2 followed by Section 6.3 which presents the dynamic connectivity of the non-wetting phase, the dynamic saturation profile of the wetting phase for the wide range of capillary numbers, and pore and throat occupancy of brine, oil and intermittency in three flow regimes. Section 6.4 describes the flow regimes that we observed and compares our observations with other results in the literature and the results obtained using laboratory-based micro-CT.

6.1 Classification of flow regimes

This chapter will study the relationship between flow rate, pressure gradient and pore-scale occupancy using X-ray imaging with a high spatial and temporal resolution. We propose a mathematical model for the rate dependence of the pressure gradient that is consistent with our measurements. A way of quantifying the flow regimes, particularly focussing on the onset of intermittent flow, from the properties of the porous medium and the imposed flow rate will be derived.

As previously introduced in Eq. 2.6, instead of flow rate, we quantify flow regimes in terms of the capillary number $Ca = \mu q_t / \sigma$ where μ is the average of the fluid viscosities, q_t is the total Darcy velocity and σ is the interfacial tension. We hypothesise that there are four flow regimes. These flow regimes are shown schematically in the upper Figure 6.1, while our results from the experiments presented in this chapter are shown in the lower Figure 6.1.

- 1) For $Ca \leq Ca^*$, the two phases flow with fixed pathways where the pressure gradient $\nabla P \sim Ca$, which is consistent with the traditional description of multiphase flow in porous media. In our experiments we find $Ca^* \approx 10^{-6}$.
- 2) For $Ca^* < Ca \leq Ca^i$, there begins to be some intermittency but Darcy's law still remains valid. In our experiments we find $Ca^i \approx 10^{-5}$.
- 3) For $Ca^i < Ca < Ca^v$, intermittent flow occurs and we see a non-linear relationship between pressure gradient and capillary number: $\nabla P \sim Ca^a$. The best fit to our data

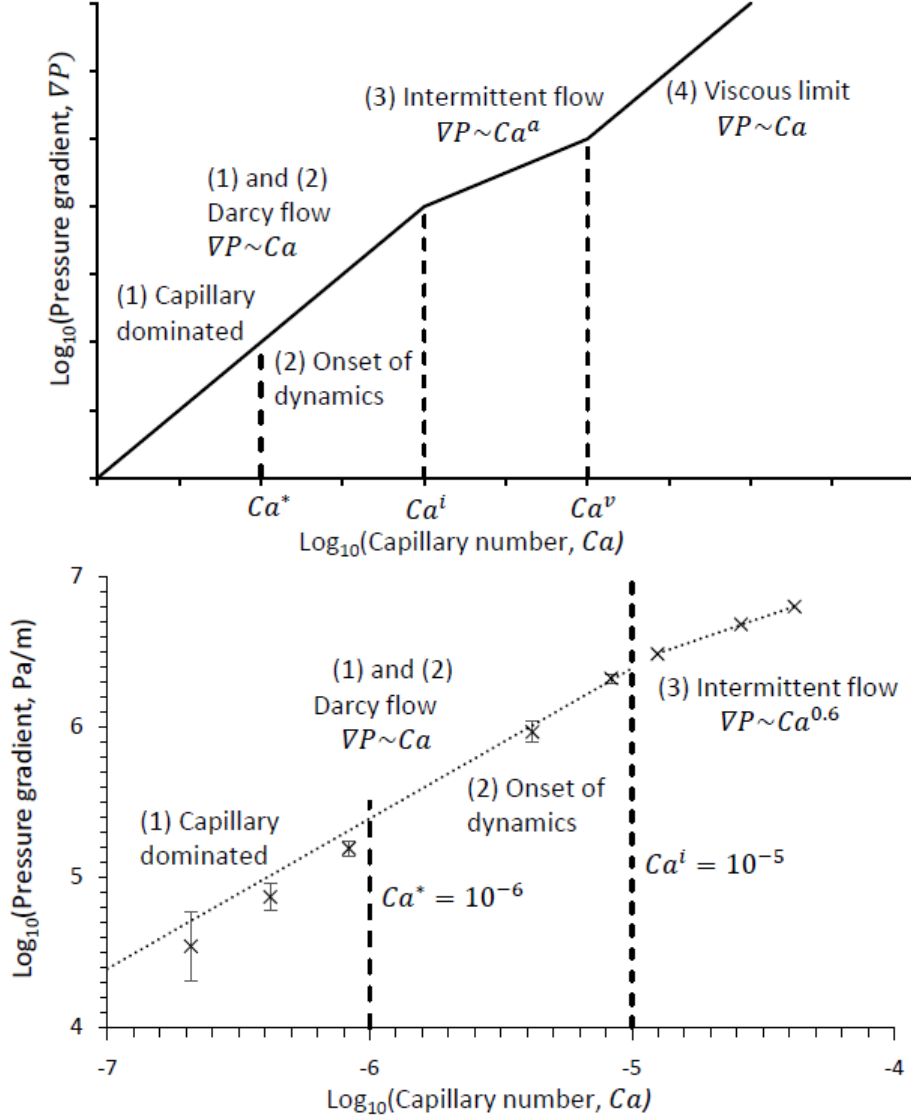


Figure 6.1: (Top figure) A schematic of flow regimes in multiphase flow. (Bottom figure) Our experimental results. In both figures, pressure gradient, ∇P , as a function of capillary number, $Ca = \mu q_t / \sigma$ is plotted on base-10 logarithmic axes. (1) For $Ca \leq Ca^*$, $\nabla P \sim Ca$ with fixed flow paths of the phases, consistent with the traditional multiphase flow in porous media, Equation (2.9). (2) For $Ca^* < Ca \leq Ca^i$, we observe the onset of dynamics, defined as the beginning of intermittency, but Darcy's law remains valid. (3) For $Ca^i < Ca < Ca^v$ intermittent flow occurs where $\nabla P \sim Ca^a$, with an empirical exponent $0 < a < 1$. (4) For $Ca \geq Ca^v$, the viscous limit is reached and again ∇P increases linearly with Ca .

gives an exponent $a = 0.6 \pm 0.1$.

- 4) $Ca \geq Ca^v$ is when the viscous limit is reached. This will not be the focus of our study, as we only consider flows where capillary forces are dominant at the pore scale.

The theoretical determination of the threshold capillary numbers presented above is intro-

duced below.

The intermittent creation and destruction of interfaces can lead to two effects: firstly, a rearrangement of fluid in the pore space, which can lead to a shift in saturation, but without changing the flow conductance for a fixed ratio of oil and water flow rates: Ca^* is defined as the capillary number at which this occurs. The second effect is when this fluid rearrangement leads to a noticeable shift in both saturation and conductance, at which Ca^i is defined, allowing the easier flow of both fluids for $Ca^i \geq Ca^*$. During intermittent flow, interfaces between the wetting phase and the non-wetting phase are formed and then destroyed to facilitate flow. The intermittent presence of conductive pathways can be analysed from a balance of interfacial energy (Reynolds et al., 2017). The energy to create an interface at the pore scale scales as σr^2 (the energy per unit area times a typical area of an interface in a single pore), while the energy added to the system because of the flow is $\Delta P \Delta V$, where ΔV is the volume change of the fluid per unit time. The pore volume is

$$\phi l^3 = \Delta V \quad (6.1)$$

where l is a typical distance between pores, ϕ is the porosity. While

$$\Delta P \sim -l \nabla P \quad (6.2)$$

over a length l , by multiplying Eq. 6.1 and Eq. 6.2, we can assume

$$\Delta P \Delta V \sim -\nabla P \phi l^4. \quad (6.3)$$

At the single pore scale, the energy added to the system equals to the energy creating the interface as

$$\sigma r^2 \sim -\nabla P \phi l^4. \quad (6.4)$$

The balance of viscous to capillary forces is quantified by the capillary number, defined here as

$$Ca = \mu q / \sigma \quad (6.5)$$

where μ is the viscosity, and σ is the interfacial tension. We assume that, Ca^i , the onset of intermittency, occurs at a capillary number at or below the value implied by Eq. 6.4. We

also know that $Ca^* \leq Ca^i$, hence substituting $\nabla P \sim -(\mu q_t)/k$ and Eq. 6.4 into Eq. 6.5,

$$Ca^* \leq Ca^i \leq \frac{-\nabla P k}{\mu} \frac{r^2}{-\nabla P l^4 \phi} \mu = \frac{kr^2}{\phi l^4}. \quad (6.6)$$

For the Bentheimer sandstone studied here, where - as we show later - we have estimates of k , r and l , this gives $Ca^i \leq 10^{-4}$ for $\phi = 0.2$.

For a single phase flow, the viscous pressure drop between two pores (a distance of l) is $\mu q_w l/k$. The local capillary pressure given by the Young-Laplace equation is approximately σ/r . Assuming a relative permeability of order 1 with $q_w \sim q_t$, then

$$\mu q_t l/k \approx \sigma/r. \quad (6.7)$$

We know viscous forces begin to dominate at the pore scale when

$$Ca > Ca^v, \quad (6.8)$$

so we can assert that, in this flow regime, using Eq. 6.7,

$$Ca^v \sim \frac{\sigma k}{\mu l r} \frac{\mu}{\sigma} = \frac{k}{l r} \quad (6.9)$$

For our data, shown in the lower Figure 6.1, the best fit to the exponent is $a = 0.6 \pm 0.1$. The measured permeability of Bentheimer sandstone is $k = 2.108(\pm 0.042) \times 10^{-12} \text{ m}^2$, the average distance between pores $l = 79 \text{ } \mu\text{m}$, and the mean throat radius $r = 24 \text{ } \mu\text{m}$ (Blunt, 2017), hence $Ca^v \sim 10^{-3}$, which is about one order of magnitude greater than Ca^i .

When $Ca^i < Ca < Ca^v$, as described by Sinha and Hansen (2012) and Sinha et al. (2017), there is a power-law scaling of flow rate with pressure drop across the system,

$$\Delta P \sim Ca^a \quad (6.10)$$

6.2 Experimental method

A series of experiments were conducted on Bentheimer sandstone with a porosity of $20.0 \pm 0.4 \%$ obtained from the micro-CT image. Oil and water were injected simultaneously

at a fractional flow of 0.5 for a series of increasing total flow rates. The core preparation has been described in Chapter 4.2. The sample was a cylindrical core 6.01 ± 0.01 mm in diameter and 57.11 ± 0.01 mm in length. A relatively long core was used to minimize the capillary end effect and only the middle part (with a length of 5.2 mm) was scanned.

To increase the contrast in the X-ray images between the wetting phase and non-wetting phase, a solution of 15 wt% Potassium Iodide (KI) brine was applied in this study. This facilitated segmentation of brine and oil phases. The viscosity of brine is 0.83 ± 0.01 mPa·s. The non-wetting phase was decane that has a viscosity of 0.838 mPa·s (provided by PubChem, open chemistry database). The interfacial tension (IFT) between brine and decane was measured to be 45.72 ± 1.76 mN/m at ambient conditions measured by a Ramé-Hart apparatus (590 F4 series) using the pendant drop method, the same method mentioned in Chapter 2.

We used a fast synchrotron X-ray micro-CT to investigate the steady-state two-phase flow behaviour at a series of capillary numbers, as listed in Table 6.1, from low flow rates representative of subsurface flow conditions to high flow rates representative of flow conditions near the wellbore. Capillary number here is defined as before, as $Ca = \mu q_t / \sigma$, where μ is the mean viscosity of the two fluids, σ is the interfacial tension between the two phases and q_t is the total Darcy velocity of the two fluids, $q_t = q_o + q_w$.

Table 6.1: List of experiments.

$Q_t(\text{mL/min})$	0.02	0.04	0.08	0.4	0.8	1.2	2.5	4
$Ca \times 10^{-7}$	2.1	4.2	8.3	42.0	83.0	130	260	420

The experimental protocol is described in Chapter 4.2. In the work presented so far – Chapter 5 – intermittency, or a fluctuating pore occupancy, is identified by an intermediate grey-scale in hour-long images. In this chapter, images are taken every 60s, and we can see directly pores where the occupancy fluctuates when Ca is larger than 10^{-6} .

6.3 Results

In this section, the dynamic connectivity of the non-wetting phase, the dynamic saturation profile of the wetting phase for the wide range of capillary numbers, and pore and throat occupancy of brine, oil and intermittency in three flow regimes are analysed and presented.

6.3.1 Dynamic connectivity of the non-wetting phase

The pore scale detail of dynamic connectivity of the non-wetting phase is shown in Figure 6.2. In this figure, the oil phase is connected through a complex pore space at low Ca , 2.1×10^{-7} . However, the non-wetting phase moves intermittently through some smaller throats occasionally at high Ca . Figure 6.2(c) shows the oil phase connected when $Ca = 8.3 \times 10^{-6}$, while it disconnected 10 minutes later, as Figure 6.2(d) shows. At the lowest flow rates, fluid configurations are governed entirely by capillary effects and remain fixed in time. We begin to see some local rearrangement of the fluids, which we call intermittency, at around $Ca^* = 10^{-6}$: see Figure 6.1.

Figure 6.3 shows the pressure differential recorded during the experiment from low to high capillary numbers. At the lowest flow rates, in the capillary-dominated regime (1), the trace is constant, indicting a fluid configuration that does not change over time, or when the flow rate is altered – these results correspond to what we observed previously, see Chapter 5. When Ca is larger than $Ca^* \approx 10^{-6}$, the onset of pressure fluctuations and pore-scale dynamics are observed in regime (2), again consistent with our previous work, Section 5.2. A significant impact on flow conductance is seen during regime (3). The intermittency then leads to an increase in flow conductance, see Figure 6.1.

We now quantify these observations by measuring the fraction of the pore space for which the fluids rearranged. We observed two types of fluctuation: the first, called type 1, is when we see a distinct change in the phase occupying a voxel from one scan to the next. This represents a change in fluid configuration that occurs over a time-scale of a minute or more. The second behaviour, called type 2, is when the fluctuations are sufficiently rapid that they occur within 1 minute. In this case we cannot explicitly see the changes; instead a voxel has an intermediate grey-scale value between water and oil. The fraction of the number of void-space voxels that are type 1 or type 2 are shown as a function of capillary number in Figure 6.4. For type 1 we measure that fraction of the voxels that change occupancy from oil to water or vice versa between two consecutive scans, averaged over the final 30 scans at each flow rate. For type 2 we simply record the fraction of intermediate grey-scale voxels. The error bars indicate the standard deviations in these measurements from image to image.

In regime 1, capillary controlled, we see no rapid fluctuations in occupancy, type 2, and

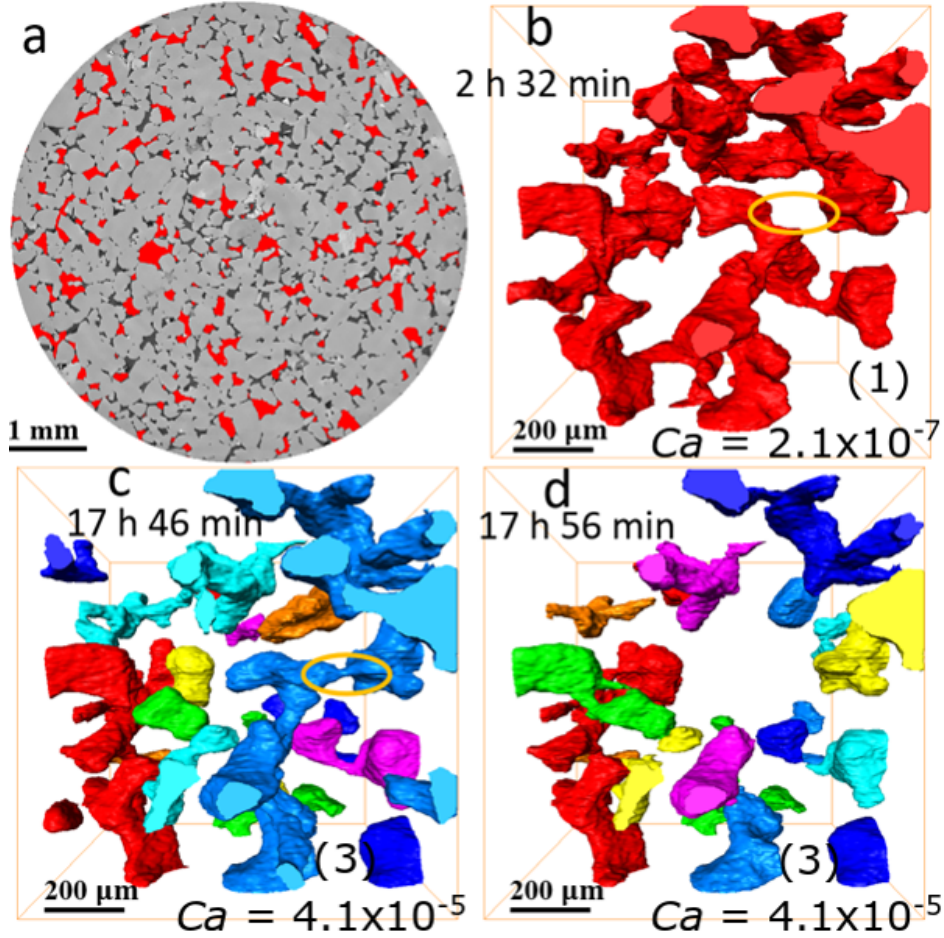


Figure 6.2: Segmented images of selected regions of the pore space. (a) A two-dimensional cross-section of a three-dimensional image in the capillary-dominated regime, $Ca = 2.1 \times 10^{-7}$, with oil shown in red, solid in grey and brine in black. (b) An example three-dimensional configuration where only oil is shown in red, for $Ca = 2.1 \times 10^{-7}$. The oil is constrained to occupy only the larger portions of the pore space and has insufficient energy to connect the two regions shown by the ellipse. (c) The same portion of the sample is shown at a higher flow rate, with intermittent flow, regime (3), where $Ca = 4.1 \times 10^{-5}$ and the oil has sufficient energy to make the short-cut to open up a more conductive flow path. The colours indicate different isolated clusters of oil. (d) This pathway is not fixed, however, and an image, taken 10 minutes later, shows that the connection has been temporarily broken.

only occasional alterations in configuration. This is illustrated in Figure 6.5 where it is evident that there are periods when there is no change in the pore-scale occupancy at all. In terms of flow and saturation we can consider that the fluids effectively move through fixed pathways. Regime 2 is characterized by more frequent type 1 rearrangements, Figure 6.5, that are seen between every scan, but there are no high-frequency fluctuations: there is no type 2 intermittency, see Figure 6.4. Here we always see some level of pore-scale dynamics, but there is no apparent impact on the linearity of the Darcy's law. In regime

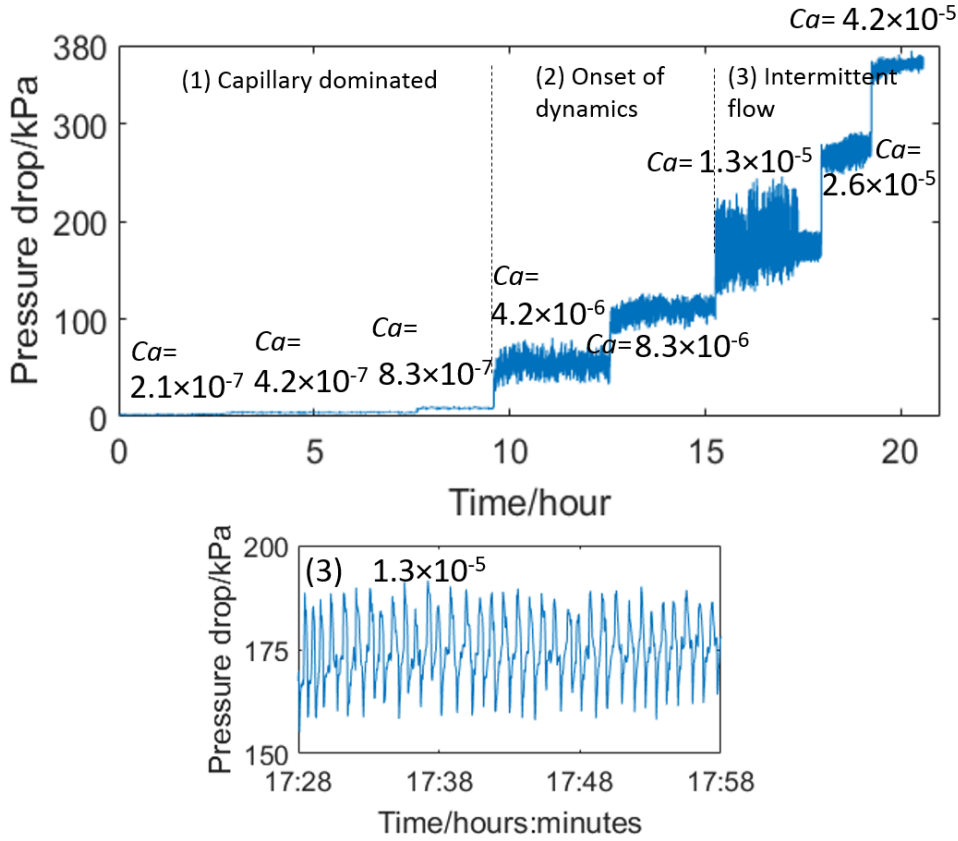


Figure 6.3: The pressure differential across the porous medium as a function of time; the capillary numbers are also indicated. The pressure drop in the flow lines has been subtracted. At the lowest flow rates, in the capillary-dominated regime (1), the trace is constant, indicating a fluid configuration that does not change over time, or when the flow rate is altered. At $Ca^* \approx 10^{-6}$ we see the onset of pressure fluctuations and pore-scale dynamics, regime (2). A significant impact on flow conductance is seen for $Ca > Ca^i \approx 10^{-5}$, regime (3). These fluctuations are of a magnitude equal approximately to a characteristic capillary pressure, indicative of local pore-scale displacement, shown more clearly in the inset.

3, when a deviation from the linear Darcy law is observed, we see the onset of rapid, type 2, changes in occupancy. These are sufficiently extensive to alter the flow conductance.

Figure 6.2 shows that oil phase configuration is different in different pore throats at each time step, and there is a dynamic connection and disconnection between oil-filled pores. This change in connectivity in regime 1 and regime 2 is illustrated in the sub-volume time sequence shown in Figure 6.6. However, in regime 3, because the intermittency (type 2) is observed, it is impossible to identify all oil phase from the images directly.

Figure 6.6 shows the interactions between ganglia population at low and high Ca respec-

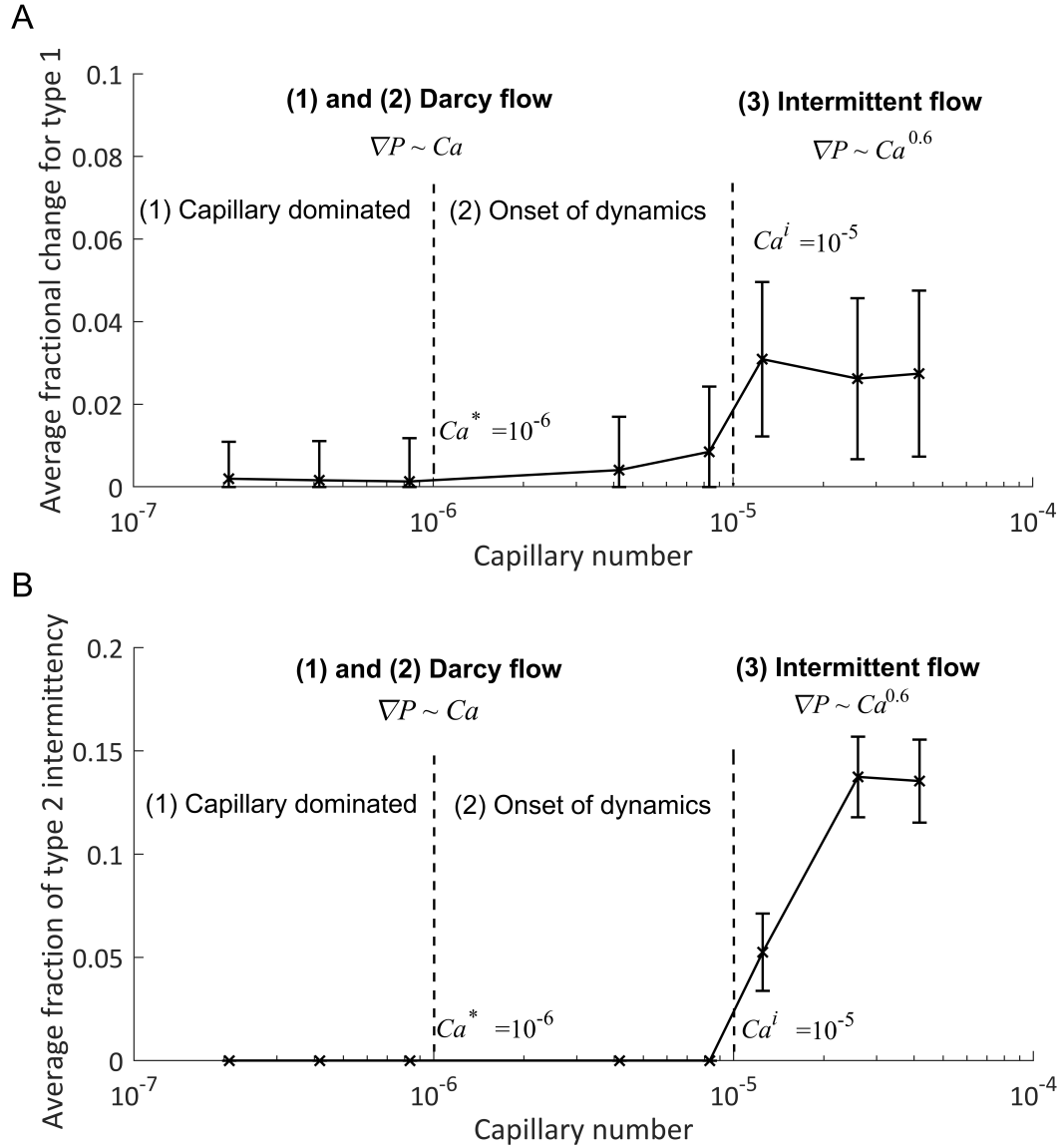


Figure 6.4: The fraction of void-space voxels for which the fluid occupancy changes from one scan to the next (type 1, A) and during a scan (type 2, B) as a function of capillary number Ca . The values are averaged over the final 30 images at each Ca . The error bars indicate the standard deviation in the values. For reference the flow regimes are indicated in brackets: regime 1 is characterized by no rapid, type 2, fluctuations and very little type 1, which only occurs now and then. Regime 2 again has no type 2 intermittency, but more sustained low-frequency rearrangements, while in regime 3 we see significant fluctuations of both types, leading to a change in flow conductance. The exponent of 0.6 is derived from the data in Figure 6.1.

tively. Figure 6.6(A) shows the saturation profile for the last 30 minutes at steady state when Ca is 2.1×10^{-7} , while Figure 6.6(B) shows the saturation profiles for the last 30 minutes at steady state when Ca is 8.3×10^{-6} . To visualize the connectivity clearly, the

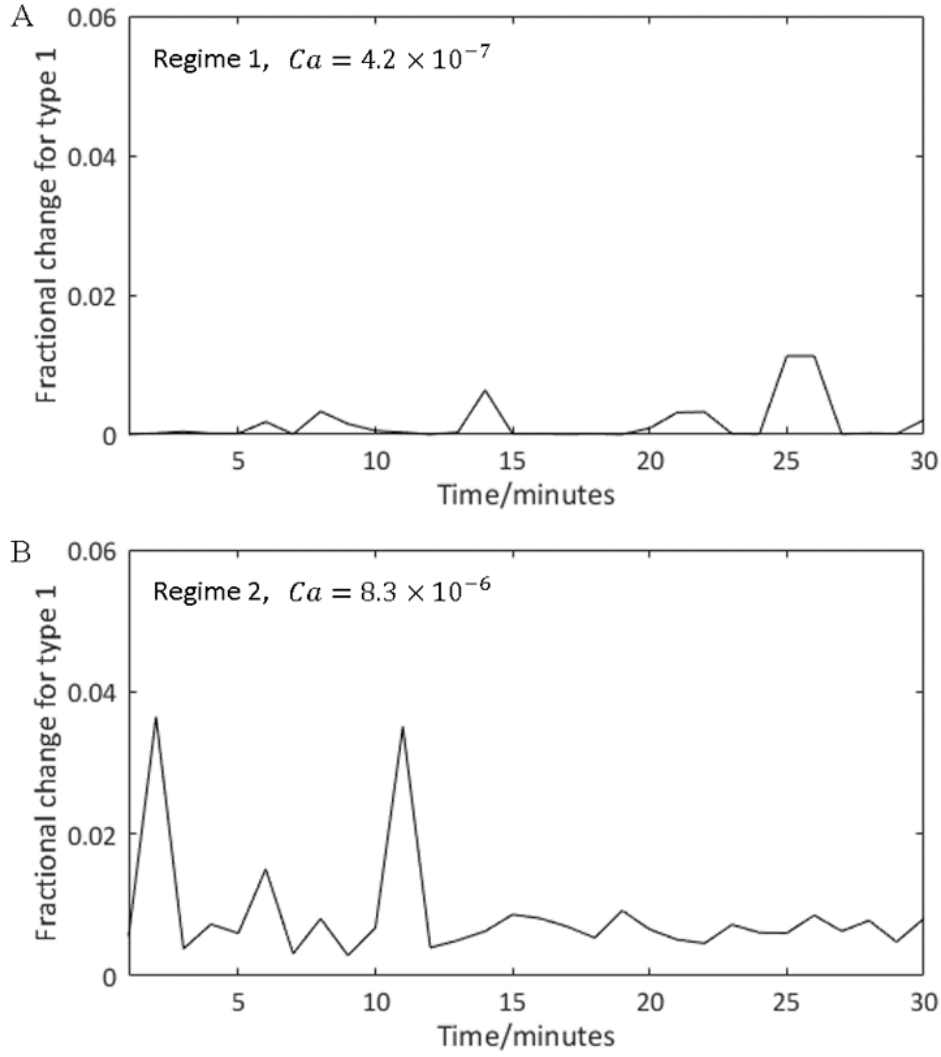


Figure 6.5: The fraction of void-space voxels for which the fluid occupancy changes from one scan to the next (type 1) as a function of time during the final half hour of the scan. (A) In regime 1, there are periods with no changes in fluid configuration. (B) In regime 2, there are always some low-frequency periodic rearrangements of the pore-space configurations from one scan to the next.

ganglia populations were subdivided into three classes and are shown in different colours in Figure 6.6(C): large ganglia, which contain thousands of pores and may be connected across the field of view (green volumes, containing more than 10^7 voxels); medium ganglia, which may be connected over tens to hundreds of pores (red volumes, containing 10^5 to 10^7 voxels); and small ganglia, which may be isolated in a single pore or be simply connected over a few pores (blue volumes, containing 5000 to 10^5 voxels). Ganglia smaller than 5,000 voxels, which had volumes less than one single pore, were ignored in this analysis. Figure 6.6(C1) shows the volume rendering of oil ganglia at a low capillary number of 2.1×10^{-7} at time 1 in Figure 6.6(A) to contrast the volume renderings of oil ganglia shown in Figure

6.6(C2) and (C3) for a high capillary number of 8.3×10^{-6} at times 2 and 3 shown in Figure 6.6(B).

We observe that at low flow rates, connected pathways of ganglia appear to be static. Moreover, the proportions of small, medium, and large ganglia stay constant over time. Again this is consistent with flow through fixed, connected pathways.

At the higher flow rates, in regime 2, the pore-scale configuration of fluids changes rapidly, as do the fractions of the oil saturation in ganglia of different size. However, in a macroscopic sense, steady-state has been reached, since the average saturations, Figure 6.6(D) are approximately constant in both space and time. If steady state had not been achieved, the entire profile would have been expected to shift with each time step. This demonstrates that, although the oil saturation remains constant, the connectivity of the non-wetting phase periodically rearranges.

The interaction between the ganglia populations observed at high flow rates is the result of alterations in dynamic connectivity. What is more, increasing the flow rate results in more disconnection and reconnection events, and the pathways have increasingly dynamic connectivity. We observe that few changes happen when Ca is less than Ca^* within the errors in our segmentation; then when Ca is larger than Ca^* some definite changes occur, while when Ca is larger than Ca^i , there are significant fluctuations in occupancy.

6.3.2 Dynamic saturation profile of the wetting phase with time

For regimes 1 and 2, water saturation was calculated based on the segmented images directly. For regime 3, the water saturation includes water that always occupied the pore and the water fraction in the intermittent flow (type 2, as defined in Section 6.3.1), which was assumed as 0.5 in this study. The water saturations calculated at each capillary number are listed in Table 6.2. Figure 6.7 shows that initially when the capillary number increases, the saturation remains constant within the uncertainty in the measurements at the lowest flow rates, which are under capillary control. However, we observe a small increase in water saturation when Ca is larger than 10^{-6} followed by a more significant change in the intermittent flow regime. If the water saturation increases, this must mean that the non-wetting phase saturation decreases: since the fractional flow is fixed, this means that the

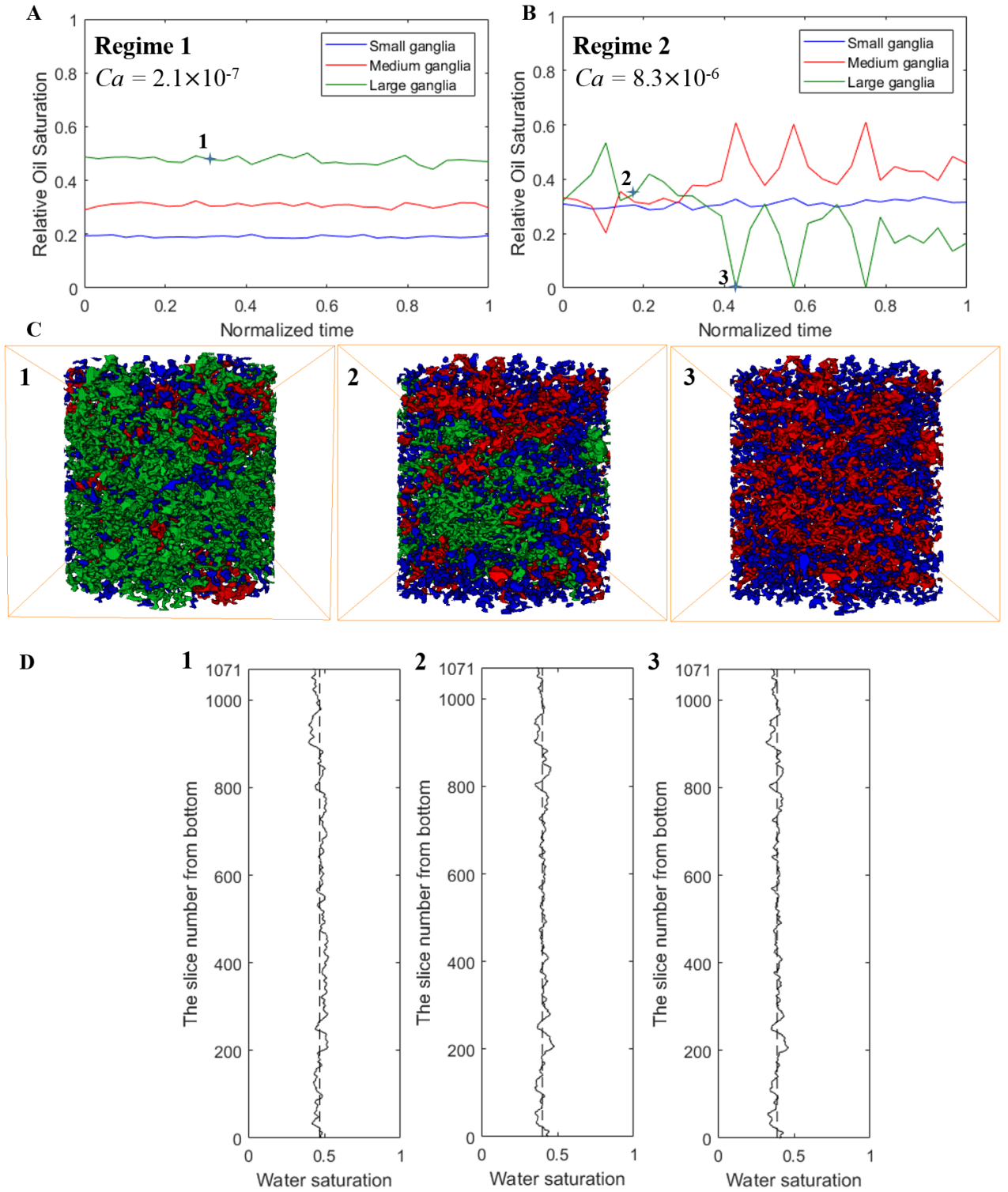


Figure 6.6: Interaction between oil ganglia populations in regime 1 and 2 without intermittency. The plots show the relative oil saturation of small (blue), medium (red) and large (green) ganglia when Ca is 2.1×10^{-7} (A) and 8.3×10^{-6} (B). (C) shows volume renderings of oil ganglia which correspond to the numbers shown in (A) and (B). Flow is from bottom to top. (D) Sliced-averaged brine saturation distribution along the sample corresponding to the numbers shown in (A) and (B).

non-wetting phase has a higher conductance at the higher flow rates, since it can sustain the same fractional flow with a lower saturation.

Table 6.2: Calculated water saturation at each capillary number.

$Ca \times 10^{-7}$	2.1	4.2	8.3	42.0	83.0	130	260	420
Mean S_w	0.53	0.53	0.55	0.56	0.59	0.61	0.68	0.70
Uncertainty of S_w	0.009	0.010	0.011	0.013	0.016	0.019	0.020	0.020

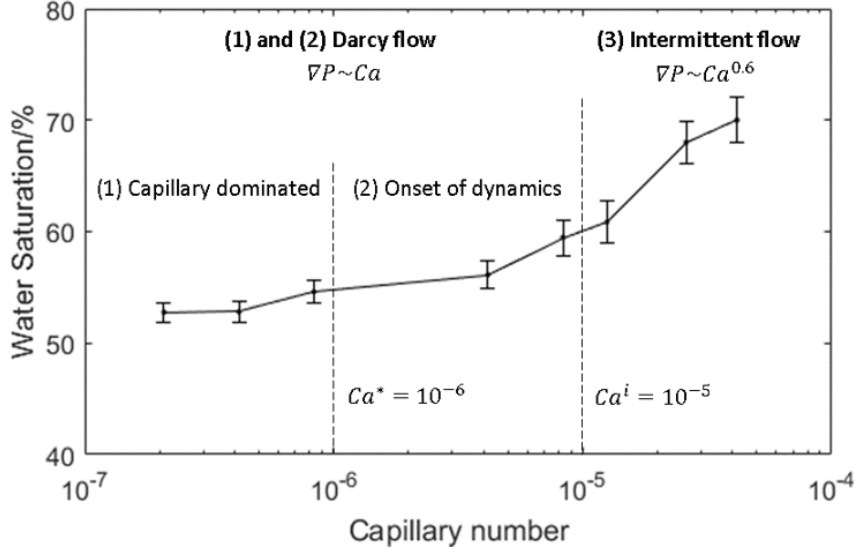


Figure 6.7: Saturation of the wetting phase as a function of the capillary number at a fixed fractional flow. Water saturation remains constant for $Ca \leq Ca^*$, increases with Ca for $Ca^* < Ca \leq Ca^i$, and further increases with a larger gradient for $Ca > Ca^i$. The uncertainty in saturation is approximately 0.015.

For each flow rate, steady state conditions are confirmed by the constant brine saturation achieved in each experiment. Brine saturations calculated from the last 30 minutes at each capillary number at steady state are shown in Figure 6.8. We see the emergence of significant fluctuations at the higher flow rates.

Combined with the results for the brine saturation at each capillary number shown in Figure 6.7 as well as the saturation during the last 30 minutes shown in Figure 6.8, we conclude that at the lowest capillary number, in regime (1), the saturation remains constant, within the uncertainty in the measurements. In regime (2), we observe a small shift in saturation and the onset of dynamic effects, with pressure fluctuations and local rearrangements of the pore space, but without a deviation from Darcy flow. At the highest flow rates in regime (3), we observe more frequent dynamic rearrangements of the pore space, as determined later from the pore-scale images.

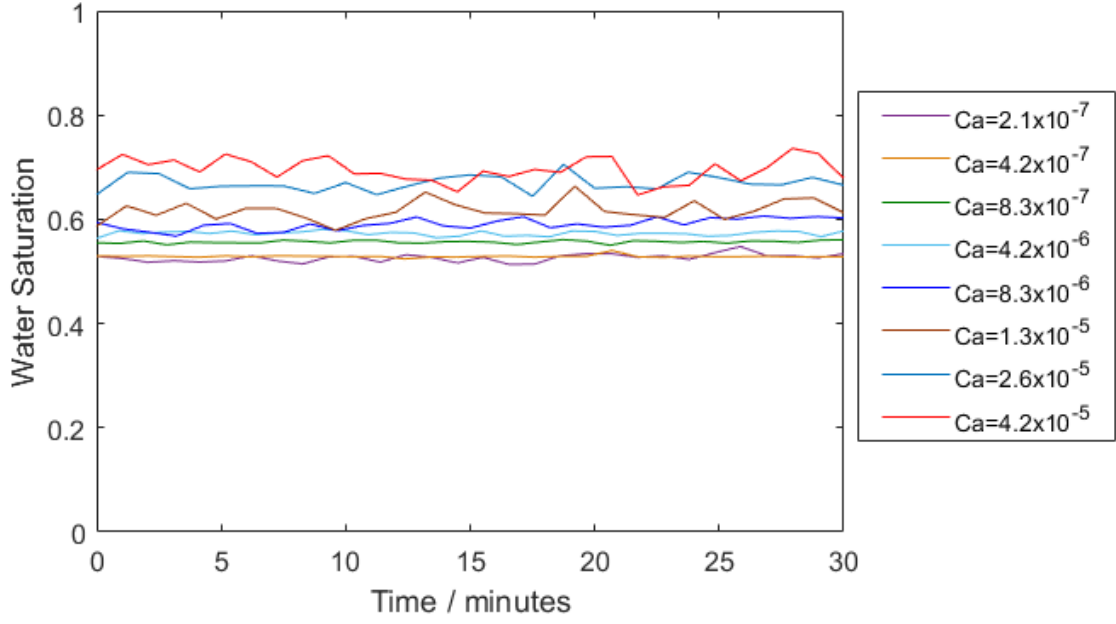


Figure 6.8: Average brine saturation is plotted versus time during the last 30 minutes at steady state at each flow rate. The brine saturation fluctuates with time. The uncertainty in the measurements is ± 0.015 .

6.3.3 Pore and throat occupancy

Generally, the oil phase occupies the larger pores because Bentheimer sandstone is strongly water-wet (Blunt, 2017). The location of the pore and/or throat, where dynamic connectivity of oil ganglia occurs and their size are investigated in this section. A generalized pore network extraction algorithm (Raeini et al., 2017) was applied to subdivide the void-space into pores representing wider regions and throats representing narrow restrictions based on the radii of each divided geometry. These voxels, whose centres were filled with brine, oil or intermittency, were used to compute the volume fraction of pores and throats. Furthermore, by computing pore-by-pore occupancy over all experimental images taken at the last 20 minutes at steady state, the standard deviations were calculated for each image. Then the average standard deviation in individual pore occupancy was obtained and plotted against the pore/throat sizes. This reflects the amount of variation at different pore/throats sizes during the last 20 minutes of steady-state for each Ca .

As evident in Figure 6.9, as expected for a water-wet medium, the oil is found in the large pores while brine resides in the corner of large pores, small pores and throats (Bultreys et al., 2018). When the capillary number was low, the oil and brine phases remained in

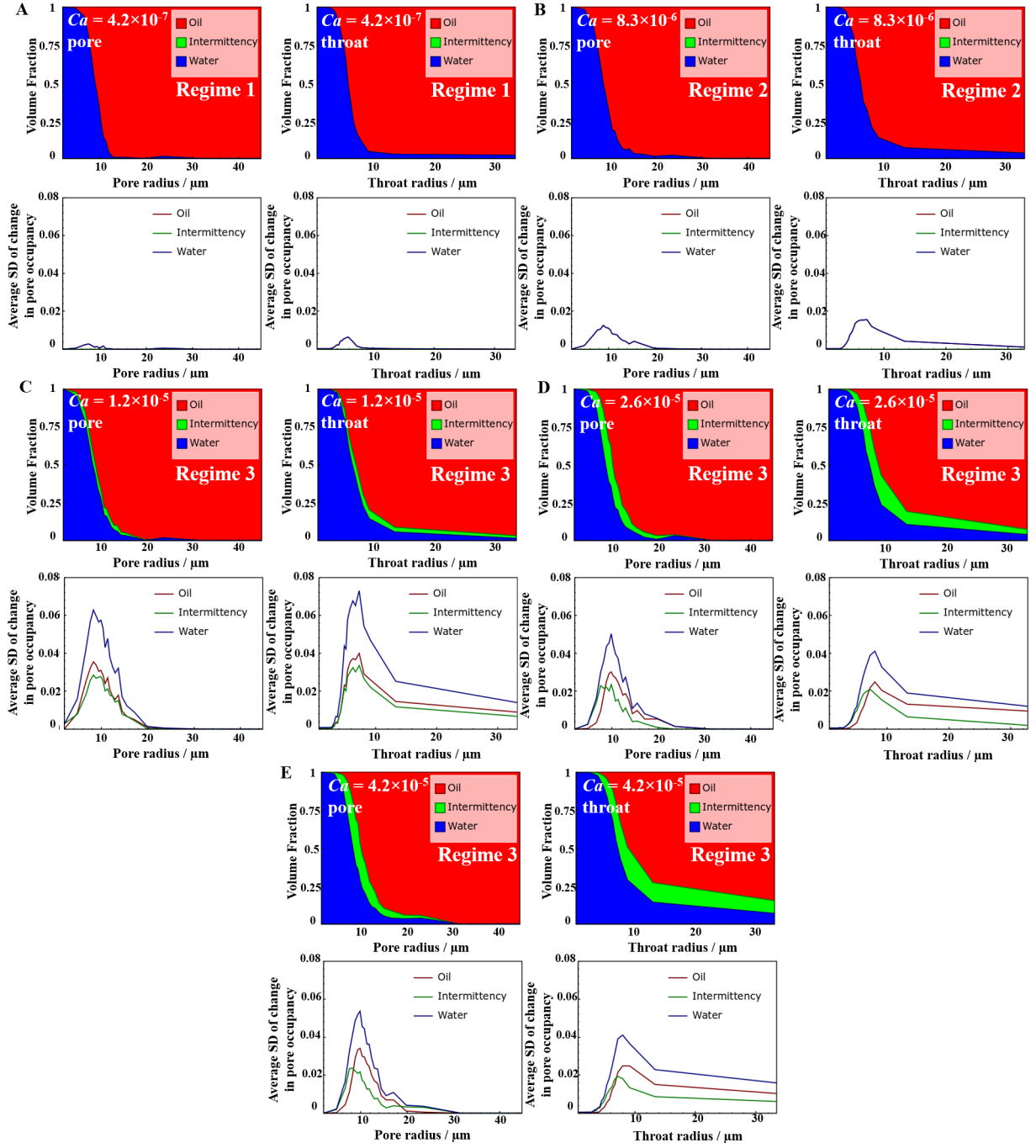


Figure 6.9: The volume fraction of radii of pores and throats occupied by oil, brine and intermittency as a function of radius when Ca is 4.2×10^{-7} , 8.3×10^{-6} , 1.2×10^{-5} , 2.6×10^{-5} and 4.2×10^{-5} . Red represents oil, green is the intermittency (type 2), and blue is brine. The lower figures show the standard deviation of change in individual pore occupancy during the last 20 minutes of steady state flow, indicating the amount of type 1 intermittency – see Figures 6.4 and 6.5.

their own fixed pathways and little change occurs. In regime 2, for $Ca > 10^{-6}$, we begin to see more significant changes in occupancy of type 1 in mainly the smaller pores. When

the capillary number is increased to 10^{-5} , these changes are more significant and occur in pores and throats of intermediate and small size. When the capillary number is very high, larger than 10^{-5} in the intermittent flow regime 3, there are regions of the pore space where – during the period of the scan - sometimes oil is present and sometimes brine, Figure 6.2. This shows that the flow pathways are not fixed, but that a dynamic or intermittent connectivity is observed. Compared with the pore occupancy at low capillary numbers, intermittent flow occurs in a larger range of pore/throat sizes at higher capillary numbers: indeed we now see changes in some of the larger pores as well.

6.4 Discussion

We now compare our results with the work described in Chapter 5. In Section 5.2, we injected both oil and brine in Bentheimer sandstone using laboratory-based micro-CT at two capillary numbers: 3.0×10^{-7} and 7.5×10^{-6} . Each scan took about one hour at a voxel size of $6 \mu\text{m}$. For the low capillary number of 3.0×10^{-7} , we observed connected and stable pathways. The water saturation was 0.52 when the fractional flow was 0.5. For the high capillary number of 7.5×10^{-6} , we observed intermediate grey-scale regions occupied by both oil and water, called intermittency, during a one-hour scan. When the fractional flow was 0.5, the water saturation was 0.56 and the percentage of intermittency in macro pores was 33%.

In this chapter, at the low capillary number of 2.1×10^{-7} , two-phase flow had fixed pathways with no intermittency, consistent with our micro-CT results, while the water saturation was 0.53, similar to 0.52 seen for the lab-based micro-CT at a similar capillary number. At a high capillary number of 8.3×10^{-6} , we saw a distinct change in the phase occupying a voxel from one scan to the next. The corresponding water saturation at this capillary number was 0.59, similar to the 0.56 seen in the lab experiments. By summing all changes of pore occupancy from one scan to the next which occurred in one hour, see Figure 6.5, the percentage of the change in phase occupancy within one hour was 26%.

The percentage of fluid rearrangement from Figure 6.5 using the synchrotron is 7% lower than that obtained using laboratory-based micro-CT. This is not a big difference, and there might be two reasons for this. First, these two experiments were conducted on two different

samples. Although Bentheimer sandstone is comparatively homogeneous, the pore structure and petro-physical parameters vary between samples. Second, the procedures of these two experiments were different. In the laboratory-based micro-CT experiment, more brine was injected as the fractional flow was increased at a fixed flow rate, see Chapter 4.1, until steady state was reached. However, in fast synchrotron micro-CT, the flow rates of oil and brine were increased with a fixed $f_w = 0.5$; hence the flow rate was increased.

In conclusion, the flow regimes identified using fast synchrotron images are consistent with the results we observed from a lab micro-CT scanner.

To continue a quantitative analysis of flow regimes to compare with other experiments in the literature, we now define wetting- and non wetting-phase capillary numbers (Datta et al., 2014a),

$$Ca_w = \frac{q_w \mu_w}{\sigma} \quad (6.11)$$

and

$$Ca_{nw} = \frac{q_{nw} \mu_{nw}}{\sigma} \quad (6.12)$$

respectively, where q is the Darcy velocity [m/s], μ is the dynamic viscosity [Pa·s], and σ is the interfacial tension between the wetting and non-wetting phases [N/m].

We vary capillary numbers over a range of Ca_w and Ca_{nw} , as shown in Table 6.3, and summarize our observations using the state diagram shown in Figure 6.10. At very low flow rates, we observe that the oil flows through an unchanging, connected 3D pathway for small values of Ca_w and Ca_{nw} , shown by the red symbols; however, as either Ca_w and Ca_{nw} increase, this pathway begins to intermittently break up, shown by the blue symbols. At the largest values of Ca_w and Ca_{nw} , the oil continually breaks up into discrete ganglia, shown by the green symbols.

Table 6.3: Wetting and non-wetting phase capillary numbers for the experiments presented in this chapter.

$Q_t(\text{mL/min})$	0.02	0.04	0.08	0.4	0.8	1.2	2.5	4
$Ca \times 10^{-7}$	2.1	4.2	8.3	42.0	83.0	130	260	420
$Ca_w \times 10^{-7}$	1.1	2.1	4.2	21.2	41.8	65.5	131.5	211.5
$Ca_{nw} \times 10^{-7}$	1.0	2.1	4.1	20.8	41.2	64.5	128.5	208.5

As shown in Figure 6.10, the first class of connected-to-broken up transition occurs when Ca_w exceeds a threshold value, $Ca^* \approx 10^{-6}$, and the second class transition is estimated

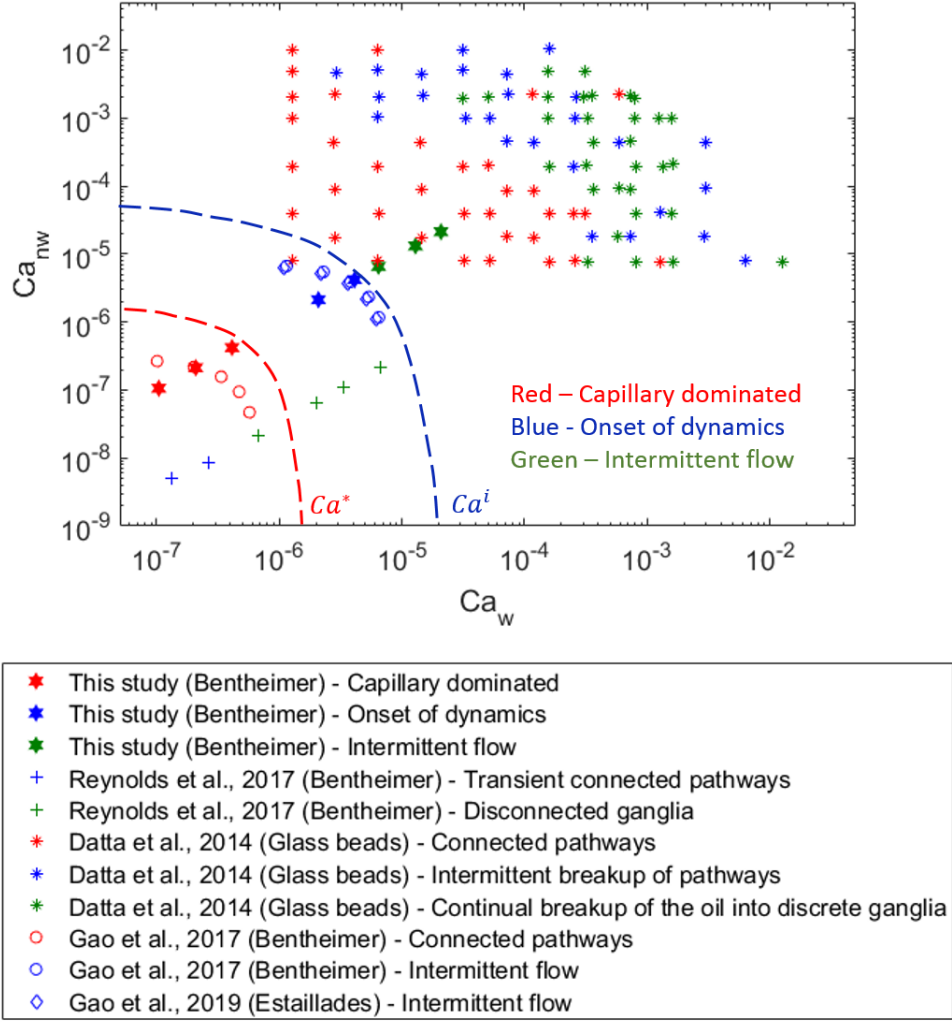


Figure 6.10: State diagram of the transition from flow with stable pathway to flow with dynamic connectivity for simultaneous two-phase flow through Bentheimer as a function of Ca_w and Ca_{nw} . Red represents regime 1 (capillary controlled). Blue represents regime 2 (onset of dynamics). Green represents regime 3 (intermittent regime). More details can be found in Table 6.3 and Table 6.4.

as $Ca^i \approx 10^{-5}$ where $Ca_w/Ca_w^i + Ca_{nw}/Ca_{nw}^i \sim Const.$ is summarized from this study. We compared our results with work from Datta et al. (2014a) and Reynolds et al. (2017). Datta et al. (2014a) performed brine-oil flow in packs of glass beads at capillary numbers 10^{-6} to 10^{-1} . Reynolds et al. (2017) conducted brine- N_2 flow in Bentheimer at capillary numbers 10^{-8} to 10^{-5} . Details of these experiments are listed in Table 6.4.

As shown in Figure 6.10, it is clear that the critical capillary numbers from Datta et al. (2014a) are larger than those from our analysis, while our critical capillary numbers are larger than those obtained from Reynolds et al. (2017). The difference in critical capillary numbers in these experiments could be due to different pore structures in a beadpack and

Table 6.4: Comparison of Ca values between this work and previously-published studies.

Author	Fluid system	Sample	Porosity	Ca_w range	Ca_{nw} range
This work	Oil-brine	Bentheimer	0.20	$10^{-7} - 10^{-4}$	$10^{-7} - 10^{-4}$
Datta et al. (2014)	Oil-brine	Bead pack	0.41	$10^{-6} - 10^{-1}$	$10^{-5} - 10^{-1}$
Reynold et al. (2017)	N ₂ -brine	Bentheimer	0.19	$10^{-7} - 10^{-5}$	$10^{-8} - 10^{-6}$
Gao et al. (2017)	Oil-brine	Bentheimer	0.22	$10^{-8} - 10^{-6}$	$10^{-8} - 10^{-6}$
Gao et al. (2019)	Oil-brine	Estailades	0.28	$10^{-7} - 10^{-6}$	$10^{-7} - 10^{-6}$

sandstone (Bijeljic et al., 2013b) and different fluid viscosity ratios. This means that we need to extend our analysis to account for different porous media and viscosity ratios.

The critical capillary numbers shown in Figure 6.10 are lower for more heterogeneous media (Estailades carbonate and Bentheimer sandstone) and higher in the more homogeneous beadpack. The critical capillary number is also lower if gas is the non-wetting phase (compare our data on Bentheimer for oil and water with Reynolds et al. (2017) for nitrogen and water in Figure 6.10). This shows that viscosity ratio also has an impact on the transition between flow regimes.

6.5 Summary

To extend the work of Reynolds et al. (2017) who studied a N₂/brine system, we investigated the oil/brine flow for a wider range of capillary numbers from 2.1×10^{-7} to 4.2×10^{-5} in this study. We have demonstrated how synchrotron imaging combined with steady-state flow can be used to identify and explain three flow regimes in detail. Co-injection micro-core floods using n-decane and brine were performed to investigate two-phase steady-state flow behaviour at a series of flow rates and three types of flow behaviour were identified from the results. At the lowest flow rates, when the capillary number is less than $Ca^* \approx 10^{-6}$, the traditional multiphase Darcy's law is valid, and both phases follow fixed pathways. When capillary number increases to $Ca^* \approx 10^{-6}$ there are some low-frequency changes in fluid occupancy, and oil saturation decreases slightly at fixed fractional flow while the total mobility is approximately unchanged. The onset of ganglion dynamics was captured and imaged within this range of capillary numbers but Darcy's law is still obeyed. When the capillary number increases beyond a critical value $Ca^i \approx 10^{-5}$, intermittent flow leads to an increased overall flow conductance at steady state and the relationship between capillary

number and pressure gradient becomes non-linear. We also observe the onset of rapid and extensive fluctuations in pore occupancy with a time scale less than the minute-long scan time. Because of the dynamic connectivity, there is interaction between oil ganglion populations, but the saturation profiles are constant at steady state.

We compared our results obtained by fast synchrotron tomography with the results obtained by the micro-CT at similar capillary numbers - these results have already been shown in Chapter 5. At low capillary numbers, it is evident that two-phase flow follows fixed pathways with no intermittency in both synchrotron and micro-CT experiments. At high capillary numbers, intermittent flow is present, and the percentages of the change in phase occupancy obtained from both experiments are similar, with 7% difference. These results indicate that the flow regimes identified using fast synchrotron images are consistent with the results we observed from a lab micro-CT scanner. Furthermore, our observations and results were compared with the results provided by Datta et al. (2014a) and Reynolds et al. (2017). The relationship between the capillary numbers over a range of Ca_w and Ca_{nw} and flow regimes was summarized on a state diagram. Our classification of three flow regimes fits the other data in the literature, although the critical capillary numbers delineating the regimes are different.

Future work could study micro-porous carbonates by fast synchrotron tomography, different fluid viscosity ratios, the full range of fractional flows and different wettabilities, including mixed-wet media (Lin et al., 2019). We suggest that the same flow regimes will be seen for different porous media, albeit with different threshold capillary numbers, as seen in Figure 6.10. In mixed-wet media, intermittent flow may be more significant than in the water-wet rock studied here (Zou et al., 2018). Accurate pressure measurements during steady-state flow, combined with pore-scale imaging, should allow an accurate and robust characterization of multiphase flow under the full range of conditions likely to be encountered in the subsurface.

Chapter 7

Conclusions and future work

7.1 Conclusions

This thesis demonstrates a repeatable and efficient methodology for measuring relative permeability and corresponding pore occupancy in both sandstone and heterogeneous carbonate samples at the mm-scale. This method combines experiment, pore-scale imaging and analytical methods together and provides a reliable way to observe how immiscible fluids flow in three-dimensional rock samples. This research has established a platform for the study of multiphase flow during simultaneous steady-state flow at the pore scale. Furthermore, our method potentially is faster, as it takes less time to achieve steady state in a smaller core, while providing pore-scale information on fluid configurations. Lastly, the quantitative analysis of pore occupancy could be used for validation or input into simulation studies to explain and predict the observed displacement behaviour.

The three types of experiments were performed:

- 1) Laboratory-based micro-CT imaging of two-phase flow at different fractional flows at both low and high capillary numbers in Bentheimer sandstone.
- 2) Laboratory-based micro-CT imaging of two-phase flow at different fractional flows at both low and high capillary numbers in micro-porous Estailades carbonate.
- 3) Fast synchrotron dynamic imaging of two-phase steady-state flow at equal volume frac-

tions of two immiscible fluids in Bentheimer sandstone from low to high capillary numbers.

Firstly, an X-ray tomography-based experiments and image analysis were performed on a Bentheimer sandstone core with a spatial resolution of $6\ \mu\text{m}$. Oil and brine were injected at different fractional flows and one-hour scans were taken at steady state. The imbibition relative permeabilities and pore occupancy at low and high capillary number were measured using a differential imaging method. By obtaining the differential image between a doped brine image and the scans taken at each fractional flow, an intermittent flow was identified from the hour-long scan which was taken at a high capillary number. Then the fraction of oil and brine in the intermittency was quantified through the grey-scale histogram distribution. When capillary number was low, the oil and brine phase occupancy was constant with fixed flow pathways through the pore space, which is consistent with conventional theory. The measured relative permeabilities compare well with literature measurements on larger cm-scale cores, which means that this novel method could be potentially used in the oil industry for larger-scale modelling. At the higher flow rate, a significant fraction of the pore space contained an intermittent flow where – during the hour-long period of the scan – the pore space was occupied for some time by brine and for some time by oil.

Next, this experimental and image analysis method was further developed to be a more general method for measuring relative permeabilities and pore occupancy. Steady-state waterflood experiments at different fractional flows were performed at a high capillary number on Estailades carbonate with a spatial resolution of $3.8\ \mu\text{m}$. In this study, the differential imaging method plays an important role in distinguishing macro-pore space, explicitly resolvable in the image, and micro-porosity, together with fluid occupancy. Four phases were classified to quantify oil and brine saturation: (i) solid grain, (ii) micro-pore space I which is occupied only by brine, (iii) micro-pore space II which can be filled with oil, brine and partially saturated by oil and brine at different fractional flows, and (iv) macro-pore space, which itself can contain oil, water or can contain an intermittent flow, where the occupancy fluctuated between oil-filled and brine-filled, at high capillary number. We also inferred initially higher recovery from micro-porosity which consists of smaller pores by calculating the oil recovery from macro-porous and micro-porous space. This is expected in a water-wet rock where the wetting phase preferentially fills the smaller pores. Then we compared the relative permeability curves between Bentheimer and Estailades. Due to the large amount of micro-porosity of Estailades, connectivity of the pore space was enhanced

and a higher end-point water relative permeability and a lower residual oil saturation were reached. What is more, there is a considerable shift to higher brine saturations because brine occupies micro-pores in Estailades.

Finally, we performed this two-phase flow (oil-brine) experiment at a synchrotron imaging facility with faster scan time to confirm, directly, this intermittent or dynamic connectivity, as seen already for brine-nitrogen flow (Reynolds et al., 2017). Co-injection micro-core floods using n-decane and brine were performed on a Bentheimer sandstone to investigate two-phase steady-state flow behaviour at a series of flow rates at the pore-scale with a spatial resolution of $5.2 \mu\text{m}$ and a temporal resolution of 60 s. Three flow behaviours were classified from the fluid rearrangement and the pressure differential. (1) The capillary-dominated flow regime, where both the wetting phase and non-wetting phase flow through fixed pathways consistent with traditional Darcy' law. (2) The onset of dynamics at $Ca^* = 10^{-6}$, where some fluid low-frequency rearrangement occurs in the pore space, and the oil saturation decreases but the pressure differential still has linear relationship with flow rate. (3) Intermittent flow for $Ca^i > 10^{-5}$, characterized by rapid fluctuations of the pressure differential and a power-law dependence $\nabla P \sim q^a$ with an exponent $a \approx 0.6$. Also, the pore scale rearrangement occurs more frequently with a time-scale less than a minute. These fluctuations periodically open short-cuts for the flow when oil has sufficient energy for a more conductive flow path at high capillary number in regime (3), which led to enhanced flow. We demonstrated that fluid rearrangement happened in pores and throats of intermediate to small sizes based on a generalized pore network extraction algorithm. Because of this dynamic connectivity, there is interaction between oil ganglion populations, but the saturation profiles keep constant at steady state.

7.2 Future work

The key application of this work is to provide an effective method to measure relative permeability and pore-scale flow behaviour in a wide range of rock types using X-ray imaging methods. Furthermore, it can be applied to many other fields of research in both single and multiphase flow in porous media. For instance, this experimental technique is readily applicable to a mixed-wet carbonate, in which micro-porosity plays an important role (Alhammadi et al., 2019b,a). Future work could include a wide variety of the pore-scale

multiphase flow problems, including those at the conditions representative of subsurface aquifers, oil and gas fields and other deep geological systems. More details will be introduced in next section.

7.2.1 Future work on two-phase relative permeability and capillary pressure

7.2.1.1 Capillary end effect and capillary pressure

Capillary pressure and the relative permeability of each phase are the two most important parameters to quantify fluid flow at the macroscale (Blunt, 2017; Herring et al., 2013; Parker, 1989). However, there are always limitations in the use of these parameters to predict subsurface fluid flow. Using conventional methods, it is difficult to measure both parameters simultaneously on the same sample. Separate measurements are always made on different samples with different flooding protocols. Thus it is not certain that the pore structure and displacement processes used to determine relative permeability are the same as those when capillary pressure was measured. Pore-scale imaging has provided accurate descriptions of fluid distribution. The capillary end effect, which is inevitable in laboratory experiments, caused by capillary pressure discontinuities at the inlet and outlet ends of the rock should be considered in our analysis (Lin et al., 2018).

One issue not fully addressed in the thesis is the inaccuracy of the predicted relative permeability caused by the capillary end effect. In our experiment, we used a long core (5 - 6 cm long) to diminish the effect caused by this capillary end effect: in the section imaged, the saturation profile was, on average, constant at steady-state. However, we mentioned the pressure drop across the whole sample where the saturation profile may not have been homogeneous (Lin et al., 2018). The relative permeabilities are firstly determined from the experimentally measured pressure drop and average saturation obtained by imaging. Then capillary pressure gradient along the whole sample is considered to correct the brine relative permeabilities. The curvature of the fluid menisci, which can be calculated from segmented images, can be used to estimate the local capillary pressure using the Young-Laplace equation (2.2). If there is a gradient in capillary pressure, this can be quantified. This enables the capillary pressure to be found directly from images, while correcting analytically for

the relative permeability.

7.2.1.2 Measurements of relative permeability and capillary pressure on a wettability-altered sample

As a massive resource of oil and gas reserves, it is always important to accurately describe the complex pore structure and flow behaviour in carbonate reservoirs. Generally, carbonate reservoirs have structural heterogeneities and are typically mixed- to oil-wet. Since wettability is one of the most important parameters controlling how fluids are arranged in the pore space (Willhite, 1986; Anderson, 1987; Blunt, 2017), how multiphase fluids behave in the complicated inner structure of carbonate should be known to make optimal production decisions.

To better understand fluid behaviour in a mixed-wet sample, Zou et al. (2018) studied drainage in an altered wettability Bentheimer sandstone and suggested that ganglion dynamics was more prevalent than in a water-wet system. Lin et al. (2019) have applied similar methods to those described in this thesis to measure relative permeability and capillary pressure during waterflooding in a mixed-wet sample of Bentheimer that had been aged in reservoir crude oil. Alhammadi et al. (2019a) extended this technique to a mixed-wet reservoir carbonate from Middle East and showed that heterogeneous structure has an impact on the fluid distribution. More work is needed to further address the impact of structure and wettability in reservoir rock systems. By applying this method, we can measure the relative permeability using X-ray micro-tomography to obtain high resolution images from which brine and oil saturation were calculated, taking into account fluid occupancy in sub-resolution micro-porosity obtained using differential imaging, while measuring the pressure drop across the sample. These images were then used to characterize mixed-wettability by measuring the in situ contact angle distribution in the pore space. Specifically, we can show how wettability and pore structure combine to provide a favourable oil recovery, with very significant implications for oil industry. All these issues need to be addressed in future work.

7.2.1.3 Future studies on two-phase flow dynamic connectivity in heterogeneous carbonates

More studies using fast dynamic tomography work at synchrotron light sources could experimentally assess the dynamic fluid behaviour of two-phase steady-state flow using the same tools and method as were used in Chapter 6. The detailed experimental methods can be found in Section 4.2.

Droplet fragmentation was described in a carbonate but was not observed in a sandstone (Pak et al., 2015). They demonstrated that droplet fragmentation process was engendered by the intrinsic heterogeneous pore structure of carbonate rocks. It would be interesting to observe how water and oil behave in both water-wet and mixed-wet carbonate samples during steady-state flow using fast synchrotron X-ray imaging. This will lead to a detailed flow regime study of carbonates to see what the differences between sandstones and carbonates.

The other main use of our work is to validate and calibrate pore-scale models: not only can we insist that these models match averaged macroscopic properties, such as relative permeability, we can test how well they predict the pore-by-pore configurations of fluids (Bultreys et al., 2018; Raeini et al., 2019).

7.2.2 More general applications

Imaging techniques combined with multiphase flow experiments are now sufficiently advanced that we can not only start to find new answers to existing questions, but also find new questions to ask. The ability to probe the pore-scale evolution of fluids and measure key petrophysical parameters could potentially provide a new and transformative way to perfect or remedy the shortcomings of the conventional and time-consuming lab experiments in the industry.

Taking advantage of the imaging techniques developed and applied in this thesis, the mechanics of tertiary flooding at the pore scale could be explained.

Core flooding experiments in the lab have proved that oil recovery can be enhanced by controlling salinity water-flooding by modifying the injected brine composition (Sheng, 2014; Jackson et al., 2016). However, how low salinity flooding increases oil recovery at

the pore-scale remains ambiguous. Mechanisms including wettability alteration (Morrow et al., 1998), mobilization of fines (Tang and Morrow, 1999; Song and Kovscek, 2015), and emulsification at elevated pH (McGuire et al., 2005) have been proposed. Among these, wettability alteration to a more water-wet state is assumed as the predominant paradigm (Sheng, 2014; Myint and Firoozabadi, 2015; Haagh et al., 2017). In order to study the microscopic mechanisms causing wettability alteration in a real oil-wet rock system, X-ray imaging techniques can help to observe and measure contact angle and curvature to see whether wettability alteration happens during low salinity flooding. By comparing the images taken before and after low salinity flooding, pore-scale details of the displacement can be obtained, including identifying the location and pore sizes where additional oil is displaced, whether the overall oil saturation decreases and whether the wettability changes occur. Thus, a systematic method combining multiphase flow experiments and micro-CT imaging can be established to study wettability alteration for a crude oil/brine system at high pressure and high temperature to enhance oil recovery.

Surfactant flooding to enhance oil recovery reduces the interfacial tension between crude oil and water phases and also changes the wettability of the reservoir rock to intermediate/water-wet. In order to observe how surfactant flooding works at the pore scale, micro-CT imaging can help to observe the curvature between oil and water phases and measure contact angle before and after surfactant flooding. Further studies on surfactant recipes can also be applied using pore-scale imaging. Pore-scale images can demonstrate the effect on EOR of various surfactant recipes. In addition to the saturation or oil recovery results obtained from core flooding experiments, pore-scale images can provide curvature, to quantify capillary pressure, and contact angle that can help evaluate the effectiveness of surfactant selected.

Polymer flooding enhances oil recovery by increasing the viscosity of the injection fluid avoiding viscous fingering and channelling to maximize sweep efficiency. Images at the pore scale can provide an effective way to follow how viscous injectant displaces the oil. Instead of evaluating the polymer quality through oil saturation at the core-scale, micro-CT images capture the whole process at the pore scale. Moreover, for a laminated sample, micro-CT imaging techniques can capture how viscous injectant flows through layers with high permeability while displacing more oil from layers with low permeability. We can follow the pore filling sequence in a real rock with the help of micro-CT, which is impossible for core-scale experiments.

Carbon capture and storage (CCS) is a vital technology that needs to be implemented globally to avoid dangerous global heating. A major concern with CCS is a long-term storage security. Micro-CT offers several advantages to assess the fate of injected carbon dioxide in the subsurface. Through providing 3D images with clear inner structure and giving good contrast between fluid and solid phases, displacement processes can be observed. CO₂ trapping in water-wet sandstones and carbonates (Andrew et al., 2014), mixed-wet carbonate (Al-Menhali et al., 2016), dynamic pore structure evolution in situ in single-mineral rock (Menke et al., 2015), and the impact of physical and chemical heterogeneity on reaction rates in multi-mineral porous media (Al-Khulaifi et al., 2017, 2019) have been studied using micro-CT. In future work, more studies can be explored on CO₂ flooding, which is one promising technique to enhance both light and heavy oil recovery which can be combined with CO₂ storage.

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