Abstract

We investigate both drainage and spontaneous imbibition processes at the pore scale using a combination of micro-fluidic experiments and lattice-Boltzmann (LB) flow calculations. First, we have fabricated a range of specifically designed etched micro-models to investigate the role of pore shape and throat width on fluid displacement. These designs include junctions with both equal and unequal channel widths in order to achieve a range of capillary entry pressures. All models were etched into Poly(methyl methacrylate) (PMMA) and chemically treated to create a hydrophobic surface. The displacement process is captured via a high-speed video microscope under ambient conditions. The experimental results were then directly compared with LB simulations. For the drainage experiments, we observe that the fluid displacement in the junction follows the Young-Laplace Law. For the case of spontaneous imbibition, however, the models with unequal channel widths display different displacement behaviour. Our experimental observations are confirmed in detail by Lattice Boltzmann Method (LBM) simulations, lending credibility to our observations. Instead of following Young-Laplace filling rules, we observe that the throat in closest proximity fills up first. This has potentially important consequences for calculation of residual saturation of CO₂ at the core scale, which is determined by spontaneous imbibition of brine following CO₂ injection.

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* Corresponding author.
E-mail address: e.boek@imperial.ac.uk
Introduction

A major concern regarding safe carbon sequestration in saline aquifers is how permanently the injected CO$_2$ will be stored underground on long time scales [1]. Four different trapping mechanisms are thought to occur during CO$_2$ sequestration: structural, solution, mineralization and residual trapping [1]. Of these mechanisms, residual trapping is of great interest since it occurs on short time scales and is driven by capillary forces. During carbon sequestration in saline aquifers, CO$_2$ is first injected in a drainage process where the (non-wetting) CO$_2$ is displacing water. Then the CO$_2$ is followed by brine in a spontaneous imbibition process as the fluids rearrange within the reservoir during the post injection stage. A fraction of the initial CO$_2$ (initial saturation) becomes trapped as a result of brine sweeping in from behind rendering the CO$_2$ immobile, which is called the residual saturation. Of particular interest are displacement mechanisms occurring at the pore scale, especially during spontaneous imbibition, for both carbon sequestration and enhanced oil recovery (EOR). Currently, Pore Network Models (PNMs) are used to predict the displacement of injected CO$_2$. For both drainage and imbibition, the displacement sequence in PNM junctions is calculated using the Young-Laplace Law [2, 3], described by

$$P_c = 2 \gamma \cos \theta (1/h + 1/w)$$  \hspace{1cm} (1)

where, $P_c$ is the capillary pressure, $\gamma$ the interfacial tension, $\theta$ the contact angle and $h$ and $w$ the height and width of the channel. For primary drainage – injection of non-wetting phase into a medium completely saturated with wetting phase - the contact angle (measured through the wetting phase) is $< 90^\circ$, resulting in a positive capillary entry pressure. This should lead to the injected non-wetting phase selecting the downstream channel with the lowest capillary entry pressure first, i.e. by entering the widest channel, as shown in Figure 1a. For imbibition – invasion of wetting phase into a porous medium initially filled with non-wetting phase – the criterion is inverted and the expectation is that the wetting phase will enter the narrowest downstream channel first, before sequentially filling all other channels in order of increasing diameter, see Figure 1b.

Figure 1. Schematic of fluid displacement sequences used in PNMs according to Young-Laplace law: a) Drainage, non-wetting phase enters widest channel first; b) Imbibition, wetting phase enters narrowest channel first.

Here we will investigate fluid displacement mechanisms in both simplified micro-models and more complex models representative of actual rock thin sections. The choice of micro-fluidic systems for these experiments is primarily driven by their transparent nature, making visualisation of fluid dynamics relatively simple. In addition to experimentally investigating the filling sequences for both drainage and imbibition, simultaneously, we compare our experiments to detailed Lattice-Boltzmann pore scale computer simulations of fluid displacement in the same topographies.
Nomenclature

\( P_C \) capillary pressure
\( \gamma \) interfacial tension
\( \cos \theta \) contact angle
\( h, w \) height and width of the capillary respectively

Materials and Methods

1.1. Materials

These models were specifically designed to explore pore geometry and the influence of varying throat diameters (30-104\( \mu \)m) for both drainage and imbibition; designs are illustrated in Figure 1. The models are fabricated in Poly(methyl methacrylate) (PMMA), (Epigem, Redcar). All have an etch depth of 50\( \mu \)m and have been chemically treated to achieve a hydrophobic surface. The range of throat diameters results in a range of capillary entry pressures given in Table 2. Both drainage and spontaneous imbibition were investigated.

Due to the hydrophobic nature of the models, \( n \)-decane (purity \( \geqslant 99\% \), Sigma-Aldrich) was selected as the wetting phase and air as the non-wetting phase. All experiments were conducted in a laminar flow cabinet (PURAIR P5-48VLF, Air Science) to limit the amount of dust coming into contact with the micro-models; all fluids were passed through a 0.22\( \mu \)m sterile filter (Millex-GP 25mm, Millipore). A high speed video microscope (12X Zoom, Navitar) equipped with a x2 magnification lens attachment (Navitar) was used to capture fluid displacement within the models with the images recorded via a FASTCAM MC2.1 camera (Photron) linked to a computer.

![Figure 2. PMMA Micro-model Designs. a) and b) have equal throat widths, c) - e) have unequal throat widths.](image)

Table 1. Capillary Entry Pressure Values (MPa) for Varying Throat Diameters

<table>
<thead>
<tr>
<th>Throat Diameter (( \mu )m)</th>
<th>Capillary Entry Pressure (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>2.19x10^{-3}</td>
</tr>
<tr>
<td>50</td>
<td>1.58x10^{-3}</td>
</tr>
<tr>
<td>64</td>
<td>1.41x10^{-3}</td>
</tr>
<tr>
<td>80</td>
<td>1.25x10^{-3}</td>
</tr>
<tr>
<td>104</td>
<td>1.15x10^{-3}</td>
</tr>
</tbody>
</table>
Methods

For the drainage experiments, two fluid pairs were investigated: de-ionised (DI) water/air and air/decane. This was the case because the DI water (non-wetting phase) in the DI water/air system was observed to be only weakly non-wetting due to the chemical treatment applied to the chips. Here the model was initially filled with air before DI water was forcefully injected in. Therefore, to observe a more strongly non-wetting system, an air/decane fluid system was investigated, where air is the non-wetting phase. Here the model was initially filled with decane (white) before air (grey) was injected into the chip at a constant flow rate.

During imbibition a single fluid pair was investigated: air/decane, though in this case n-decane is the wetting phase. Initially the micro-model is saturated with air; a droplet of n-decane is then allowed to spontaneously imbibe into the selected channel. In order to observe the effect of pore shape and throat width on fluid displacement, this was repeated for each inlet.

Simulations

We have implemented an optimized multi-phase Lattice-Boltzmann Method (LBM) [4,5] based on the colour gradient LB model introduced by Gunstensen and Rothman [6,7], to simulate two-phase flow in micro-models. This new approach uses the Multi-Relaxation-Time (MRT) collision scheme, which improves numerical stability [6] and permits higher viscosity ratio and lower capillary numbers than other multi-phase LBM models [4]. For our simulations all boundaries conditions are periodic, with a reservoir of Wetting Phase (WP) and Non-Wetting Phase (NWP) set at the bottom of the micro-model. This was done to allow imbibition of the WP and recover the NWP flowing out of the channels. The surface tension of the lattice units was set at 0.017Nm\(^{-1}\) providing stability within the simulations. A contact angle of 30° was used for all simulations, close to the experimental contact angle of 35°.

Results and Discussion

As we cannot show the videos recorded, we have reproduced here a small number of sequential still images with the aim of illustrating the entire process clearly. In addition to the experimental results, corresponding LBM simulation images are also presented for comparison.

1.2. Drainage

As can be seen in both Figures 3 and 4, drainage occurs in the order predicted by the Young-Laplace Law (Equation 1) with the largest channel becoming filled with non-wetting phase first. The LBM calculations confirm the experimental results in detail.
Figure 3. Sequence of three drainage experimental images (grey scale) for a weakly non-wetting system, including the corresponding LBM simulations (colour). The chip is initially filled with air (experimental: light grey, LBM: grey) before DI water is injected into the model (upper right, experimental: white, LBM: red).

Figure 4. Sequence of drainage experimental images for a strongly non-wet system including the corresponding LBM simulations. Here the chip is initially filled with n-decane (experimental: white, LBM: grey) before it is displaced by the injection of air (upper right, experimental: light grey, LBM: blue).

1.3. Spontaneous Imbibition – Equal Channel Throats

Figures 5 and 6 show sequences for spontaneous imbibition experiments in models having equal throat widths, with and without the inclusion of a pore. In the case of the T-junction, see Figure 5, the meniscus (thick black line) reaches the wall opposite the throat in which the n-decane is imbibing along, before traveling along the adjacent channels simultaneously. When a pore body is introduced, see Figure 6, it is observed that the n-decane will imbibe into the two adjacent channels simultaneously.

Figure 5. Spontaneous Imbibition into a T-junction with equal throat widths, n-decane imbibes from the bottom right (white) into the model which is initially saturated with air (light grey).
Figure 6. Spontaneous Imbibition into a model with equal throat widths and a circular pore geometry. \( n \)-decane imbibes from the top left (white) into the model that initially saturated with air (light grey).

1.4. Spontaneous Imbibition – Unequal Channel Throats

For the spontaneous imbibition experiments conducted in a T-junction with unequal throat widths, Figure 7, \( n \)-decane followed the displacement mechanisms as described by the Young-Laplace Law, Equation 1, with the imbibing fluid choosing the narrowest channel to travel along first.

Figure 7. Spontaneous Imbibition experiment in T-junction model, where n-decane imbibles from the top right (white) into the model that is initially saturated with air (light grey).

We observe that the addition of a fourth throat and/or a central pore body (with all throats varying in width) alters the wetting phase displacement, depending on the inlet in which the \( n \)-decane imbibes. To demonstrate that the imbibing fluid does not select the throat predicted by the Young-Laplace Law (i.e. the narrowest channel first), \( n \)-decane was imbibed into the inlet channel located opposite the narrowest channel. As can be seen in both Figures 7 and 8, the \( n \)-decane does not select the narrowest channel, but instead imbibes down the adjacent channel.

Figure 8. Still images of spontaneous imbibition of decane (upper left, white) into model with unequal throat widths that was initially saturated with air (light grey). As can be seen the imbibing decane does not enter the narrowest channel first.
Conclusions

We observe that for drainage experiments, the capillary filling rules derived from the Young-Laplace equation do accurately predict fluid displacement sequences. For imbibition, in junctions with equal channel widths (both with and without pores) the imbibing fluid responds symmetrically. However, when unequal channel widths are introduced, we observe that the capillary filling rules are not obeyed. Instead, the filling sequence depends on the pore geometry and specifically channel proximity. For this reason, current network models for spontaneous imbibition may not accurately predict fluid displacement pathways. This may have important consequences for the calculation of residual saturation of CO2 at the core scale, which is determined by spontaneous imbibition of brine following CO2 injection.

Acknowledgements

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