1. Introduction

Understanding the simultaneous flow of multiple fluids through the Earth’s crust is a fundamental problem to our society. How fluids propagate, and become trapped, governs how safely CO₂ can be stored underground in saline aquifers to mitigate climate change (Bachu, 2000; Bickle, 2009; Rubin & De Coninck, 2005), if underground drinking water supplies will be protected from spilled nonaqueous contaminants (Pye & Patrick, 1983) and how much hydrocarbon will be produced from a reservoir (Blunt, 2017). The invasion of CO₂, or the contaminant, is a transient process; macroscopic fluid properties such as saturation and pressure vary in space and time. Under steady-state conditions, the fluids have established a local equilibrium, and macroscopic fluid properties are invariant in space (Tallakstad, Knudsen, et al. 2009). However, the prevailing theory of subsurface multiphase flows depends on the equivalency of underlying pore-scale fluid dynamics during transient and steady-state flow.

The continuum framework for modeling multiphase subsurface flows assumes that each fluid phase will have its own designated pathway through the pore space, determined by capillary forces, whereby flow of that phase is independent of other fluids present (Armstrong et al., 2016; Blunt, 2017; Dullien, 1992). Thus, the fluid phase interfaces are assumed to be static, and the fluid configurations at the pore scale are in local equilibrium (Leverett, 1941; Richards, 1931). Changes to this pore-scale arrangement to accommodate changes in saturation and pressure are assumed to occur rapidly relative to the macroscopic changes. A quasi-equilibrium state at the pore scale is thus assumed, and flow properties are applied indiscriminately.
Figure 1. Schematic of (a) the current subsurface modeling framework compared to (b) a potential modeling framework applied here to study CO₂ migration and storage. The CO₂ plume shape was adapted from MacMinn et al. (2010).

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The existence of pore-scale intermittency during steady-state flow suggests the possibility of even further complexity during transient flows more typical of the subsurface. This raises the possibility that unsteady-state flows should be modeled with separate upscaled constitutive laws or flow parameters. The transition to steady state has been observed in 2-D glass micromodels as a very rapid evolution after the fluid displacement front has passed (Tallakstad, Løvoll, et al. 2009). However, more complex fluid dynamics are generally observed in rocks (Armstrong et al., 2016; Gao et al., 2019, 2020; Reynolds et al., 2017; Rücker et al., 2015; Spurin et al., 2019a, 2019b; Tallakstad, Knudsen, et al., 2009). Static interfaces have been observed between fluids within the pore space during steady-state flow when the nonwetting phase fluid viscosity is similar to or higher than that of the wetting phase, such as crude oil displacing brine (Datta et al., 2014; Gao et al., 2017; Spurin et al., 2019a).

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Using synchrotron X-ray microcomputed tomography, we imaged pore-scale fluid dynamics with high time resolution (of 1 s) while macroscopic flow transitioned from transient to steady state. We aim to establish if the pore-scale fluid dynamics underpinning larger-scale transient flow are distinguishable from the steady-state dynamics. If distinguishable, transient flows may need to be modeled by distinct flow properties from those of steady-state flows, as demonstrated in Figure 1b for CO₂ plume migration during CO₂ storage. This will require observations of the mobility of the fluid phases and the connectivity of the flow network as the fluid configuration transitions to steady state. This is explored for a nonwetting phase where intermittency would be expected at macroscopic steady state due to its low viscosity (nitrogen) and a nonwetting phase where static interfaces would be expected at steady state, again due to viscosity (decane).

The nonwetting phase (nitrogen or decane) and the wetting phase (brine) were injected simultaneously into a permeable carbonate rock initially saturated with brine. This is a drainage process as the nonwetting
Figure 2. (a) The pore space of the sample imaged prior to the injection of brine. (b) Zoom-in of the pore space shown in the white box in (a) with the maximum inscribed balls (MBs) overlaid on the pore space. (c) The pore size distribution taken from the radii of the MBs. Everything with a radii less than 2 times the imaging resolution was discarded. All pores occupied continuously or intermittently with gas are above this limit in spatial resolution.

phase initially has to displace brine to percolate across the rock sample. After this, a local equilibrium in the pore-scale fluid distribution is reached. To observe the transition to steady state initiated with two fluid phases already percolating the pore space, we also consider two further perturbations inducing unsteady state for a nitrogen/brine system, by changing the relative flow rate of the fluids.

2. Materials and Methods
2.1. Experimental Apparatus and Procedure

The experiments were conducted in a cylindrical Estaillades carbonate sample, 5 mm diameter and 20 mm length. The pore space is highly heterogeneous (shown in Figure 2a). The sample was attached to a dual injection piece, specifically designed for the simultaneous injection of two fluid phases while suppressing the generation of slug flow. The sample was initially saturated with brine (deionized water doped with 15 wt.% KI to improve the X-ray contrast). The system was pressurized to 8 MPa to minimize the compressibility of nitrogen, with an additional 2 MPa of confining pressure. Then both nitrogen and brine were injected simultaneously, with the flow rates of the nitrogen and the brine listed in Table 1. A differential pressure transducer (Keller, 300 kPa transducer with 1.5 kPa accuracy) was connected across the sample, to measure the pressure drop. Steady state was determined to be reached once the differential pressure had plateaued. The sample was resaturated with brine at the end of the nitrogen experiments (and scanned to confirm no nitrogen remained in the core), and the experiment was repeated with decane and brine injected simultaneously.

We defined steady state during an experiment using the differential pressure across the sample. For the nitrogen experiments, as discussed later, we observed oscillations in the pressure readings at steady state (see supporting information for more on the nature of the oscillations). For the initial invasion of nitrogen

<table>
<thead>
<tr>
<th>Experimental number</th>
<th>Nonwetting phase flow rate (ml/min)</th>
<th>Brine flow rate (ml/min)</th>
<th>Total flow rate (ml/min)</th>
<th>Fractional flow of brine ($f_w$)</th>
<th>Capillary number</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.015 (nitrogen)</td>
<td>0.085</td>
<td>0.1</td>
<td>0.85</td>
<td>1.6 x 10^-7</td>
</tr>
<tr>
<td>2</td>
<td>0.03 (nitrogen)</td>
<td>0.07</td>
<td>0.1</td>
<td>0.7</td>
<td>8.7 x 10^-8</td>
</tr>
<tr>
<td>3</td>
<td>0.05 (nitrogen)</td>
<td>0.05</td>
<td>0.1</td>
<td>0.5</td>
<td>5.4 x 10^-8</td>
</tr>
<tr>
<td>4</td>
<td>0.05 (decane)</td>
<td>0.05</td>
<td>0.1</td>
<td>0.5</td>
<td>2.2 x 10^-6</td>
</tr>
</tbody>
</table>

Note. Calculation of the capillary number is taken from Spurin et al. (2019a) where $Ca = \frac{q \sigma}{\rho_d \Delta r}$. For a detailed explanation of the experimental procedure, please refer to the supporting information.
Figure 3. (top) The gas saturation and differential pressure time series following the initial perturbation (Experiment 1 in Table 1). (bottom) Images of the pore space at the indicated times in Experiment 1. The gas is red, the brine is gray, and the rock grains are transparent. The first three images are for the entire imaged section of the rock. At steady state the dynamics are more subtle, and so the final three images are zoomed in on a smaller region of the pore space indicated by a square in Panel c. See the supporting information for videos of the dynamics.

into the system, the pressure increased monotonically before oscillating about a mean (see Figure 3). Steady state corresponds to the transition from the monotonically increasing pressure differential to the time where pressure oscillates around a constant mean. For the subsequent nitrogen experiments (2 and 3 in Table 1), we chose imaging windows after the time where the mean of oscillating pressure fluctuations is no longer monotonically increasing.

2.2. Synchrotron Imaging

The X-ray imaging was conducted at the TOMCAT beamline at the Swiss Light Source, Paul Scherrer Institut, Villigen, Switzerland. The sample was exposed to filtered polychromatic X-ray radiation with a peak energy of about 26 keV originating from a 2.9 T bending magnet source. The filter was 2.300 μm thick silicon. An in-house developed GigaFRoST camera (Mokso et al., 2017) and a high numerical aperture white-beam microscope (Optique Peter) with 4X magnification (Bührer et al., 2019) were used, yielding an effective pixel
size of 2.75 μm. Each tomogram contained 1,000 projections overreach 180° rotation. Each scan lasted 1 s, with a further 1 s required for the sample to rotate back to its initial position before the next scan could begin. With this temporal resolution, all interfaces were resolvable; that is, no blurring was observed as was the case in previous research (Gao et al., 2020; Reynolds et al., 2017; Spurin et al., 2019a, 2019b).

2.3. Image Analysis

Each image analyzed was 4,224 × 4,298 × 4,263 μm in size. With a sample length of 20 mm, we imaged and analyzed the middle 5 mm (this means that dynamics do occur outside the field of view in the direction of flow, but the entire cross section of the core was imaged). The images were reconstructed from the X-ray projections using the propagation-based phase contrast method (Paganin et al., 2002) and the gridrec algorithm (Marone & Stampanoni, 2012). The images were then filtered with a nonlocal means filter to suppress noise while maintaining the information at phase boundaries (Schlüter et al., 2014). The first image was taken with just deionized water in the pore space. This image is used to segment the pore space from the rock grains using a watershed segmentation algorithm (Beucher & Meyer, 1993). Then the sample was saturated with the brine, and another image was taken; this is the brine-saturated image. All subsequent images with the nonwetting phase (nitrogen or decane) and brine present were subtracted from the brine-saturated image; this results in a differential image whereby only the location of nonwetting phase remains. From this a simple grayscale value threshold can be used to segment out the nitrogen or decane. The pore space was overlain on this segmentation to locate the pore space occupied with brine.

The pore space morphology was extracted using a maximal inscribed spheres (maximum ball, MB) network extraction technique (Dong & Blunt, 2009; Raeini et al., 2017). The fluid occupancy for each pore MB was assigned for every time step. Then for every pore, the neighboring pores are listed with their occupancy. Using this, the size of an intermittent event can be calculate. Estaillades contains both macroporosity and subresolution microporosity, but the pore size range occupied by gas (either continuously or intermittently) is at least 4 times larger than the spatial resolution of the experiments, such that the microporosity is not of interest in this work (Figure 2c).

3. Results

There are four experiments described in this section. These experiments and their parameters are listed in Table 1. In Experiment 1 we observed the transition to steady state from the initial injection of nitrogen into a brine-saturated rock. After steady state had been achieved for Experiment 1, and both fluids percolate through the pore space, the system is perturbed twice by changing the flow rate of brine and nitrogen (while the total flow rate remains the same); these changes represent Experiments 2 and 3. The sample is then resaturated with brine, and the transition to steady state is observed for decane injected simultaneously with brine; this is Experiment 4. We determine macroscopic steady state primarily through observations of the average pressure gradient across the rock sample but also verify that the average fluid saturation is unchanged.

3.1. Transition to a Steady State With Intermittency

The nitrogen and brine were injected simultaneously into a rock saturated with brine (see Table 1 for experimental parameters). The flow was capillary dominated (\( Ca = \frac{q}{\sigma \lambda} = 1.6 \times 10^{-7} \) where \( q \) is the total Darcy flux, \( \sigma \) the interfacial tension between the nonwetting phase [nw] and brine, and \( \lambda \) the mobility) and the mobility \( (\lambda = \frac{\mu_{\text{nw}}}{\mu_{\text{brine}}} + \frac{1-f_w}{\mu_{\text{brine}}} \) where \( \mu \) is the viscosity and \( f_w \) is the brine flow rate as a fraction of the total flow rate) indicated that intermittency was expected at steady state (Spurin et al., 2019a). Macroscopic steady state was determined based on the differential pressure across the rock sample (Figure 3). The pressure differential increased with the initial invasion of the nonwetting phase. It then continued to increase after the nonwetting phase had percolated across the rock, before finally plateauing and oscillating around a constant mean value with a period of approximately 10 min, which was considered steady state.

Prior to percolation of gas across the sample, the pressure increased monotonically (Labels a and b in Figure 3), and there were large changes in gas saturation as gas was establishing a percolating path through the pore space. This did not induce major changes in the number of disconnected gas regions (ganglia), shown in Figure 4. The gas propagated in large bursts called Haines jumps (Berg et al., 2013; Bultreys et al., 2015; Haines, 1930) that incorporated a large number of connected pores (Figure 5).
After percolation across the sample but prior to steady state, the gas saturation increased, but less rapidly than before percolation, and the number of disconnected gas regions increased as the pathway through the pore space was optimized (Figure 4). This change in flow behavior corresponded to a decrease in the rate of change of the pressure differential in Figure 3. During the transition to steady state, the average number of pores involved in an intermittent event decreased from more than 30 pores to less than 5 (Figure 5). The total number of intermittent events also decreased (Figure 5). The number and size of intermittent events decreased as the gas found the optimal pathway through the system; the pressure still increased monotonically (Figure 3).

At steady state, the gas saturation plateaued and oscillated around an average value of $S_g = 0.26 \pm 0.02$, with a cycle lasting approximately 10 min in Figure 3; there was no net change in saturation when averaging over a differential pressure cycle. The number of disconnected gas regions fluctuated significantly (the range was $\pm 70\%$ of the mean) at steady state, but the fluctuation in the saturation was much smaller ($\pm 15\%$ of the mean) (Figure 4). The size of the intermittent events was significantly lower than during the transition, generally involving only a single pore (Figures 3 and 5). This means that small changes at critical locations in the pore space led to major changes in the connectivity of the gas, demonstrated by the disconnecting and reconnecting of a region of gas from the main flow pathway by one periodic pathway that encompasses only one pore in Figures 3d–3f.

3.2. Transition to Steady State After a Perturbation

The system was perturbed twice by changing the relative flow rates of the fluids while maintaining the total flow rate of gas and brine (Experiments 2 and 3 in Table 1). A flow pathway had been established from the initial percolation (Experiment 1), but by increasing the gas flow rate, the capillary pressure was increased and the gas was able to enter previously inaccessible pores. With this perturbation, the changes in system
properties—pressure, saturation, and intermittency—were more subtle than with the initial percolation (Figure 6).

Figure 7 shows that the majority of the pore space was continuously occupied by either brine or gas (less than 2% of the pore space was occupied intermittently by both phases). However, the impact of these intermittent flow pathways on the connectivity of the gas was significant, as shown in Figure 4. The peak for the amount of intermittent pathway flow during steady state was observed when the brine flow rate constituted 70% of the total flow rate (Experiment 2 in Figure 7, with details in Table 1). This agrees with previous observations of intermittency during steady-state flow (Spurin et al., 2019b). We therefore observed the transition to a state with more intermittent pathway flow (from Experiment 1 to Experiment 2) and, subsequently, the transition to a state with less intermittent pathway flow (from Experiment 2 to Experiment 3).

When transitioning to steady state for Experiment 2, where the steady-state intermittency is more than the previous step, we observe a gradual increase in gas saturation and intermittency (Figures 6 and 7, respectively). This makes this transition markedly different from the initial percolation (Experiment 1), where the dynamics were largest at the onset and then reduced to small movements (Figure 5). Many of the flow pathways were established during the initial percolation phase, and so the transition to steady state was quicker. The modest increase in intermittency is associated with the opening of new pathways to accommodate the greater gas flux. The long-period (approximately 10 min) pressure differential oscillations persist through the perturbation (Figure 6).

When transitioning to steady state for Experiment 3, where the steady-state intermittency is less than the previous step, we observe an increase in gas saturation and a gradual decrease in intermittency (Figures 6 and 7, respectively). The transition to steady state is quickest for this experiment (Figure 6).

Overall, transitions to steady state after an initial percolation had occurred are characterized by gradual changes in intermittency between an initial and final saturation state regardless of whether the intermittency is increasing or decreasing. However, even with little change in saturation the size of intermittent events is much larger during the transient than steady-state flow, similar to observations made during the initial percolation (Figure 8). It is thus a general feature of flow during both the initial percolation and subsequent perturbations that unsteady-state movements incorporate larger events (up to 20 pores involved in

Figure 6. The gas saturation and differential pressure as a function of time. The raw pressure data are shown in light coloring, while the bold coloring shows the pressure data averaged over a period of an oscillation to clearly show the transition to steady state. See Table 1 for the experimental parameters.

Figure 7. The percentage volume of the pore space identified as intermittent. Each point is the average of a 10 min window of continuous imaging. The different experiments (as listed in Table 1) are labeled on the image.
Figure 8. The size of an intermittently occupied part of the pore space decreases as we approach steady state for Experiment 2, where a perturbation was initiated in a steady-state percolating system by changing the relative flow rates of nitrogen and brine. Figure 5 is the equivalent figure but for Experiment 1. Similar evolutions are observed for Experiments 3 and 4. Each analysis spanned a 30 min imaging window.

The initial transition to steady state was much quicker (taking less than an hour) than with gas, shown in Figures 6 and 7. The oil saturation is higher than the gas saturation in Experiment 3 as expected from increasing the nonwetting phase viscosity (Figure 6). The fluctuations in pressure are much smaller than for the gas experiments. We do observe intermittency during the transition to steady state. However, it is far less prevalent than during the initial nitrogen percolation (Figure 7), and it evolves more quickly (Figure 6). As expected, there is almost no intermittency at steady state (0.2% of the pore space is identified as intermittent).

3.3. Transition to a Steady State With No Intermittency

The sample was resaturated with brine, and an experiment was conducted with decane as the nonwetting phase to explore the role of viscosity on the transition to steady state (Experiment 4 in Table 1). The viscosity ratio has been identified as a primary variable controlling intermittency, with less dynamics expected at steady state for systems with more viscous nonwetting fluids (Spurin et al., 2019b). The flow rate was matched with the gas experiments, and the capillary number is higher by an order of magnitude due to the viscosity of the oil (although flow is still capillary dominated, Table 1).

4. Conclusions

Intermittent fluid flow pathways were always observed during macroscopically transient flow. These intermittent flow pathways constituted a larger volume of the pore space and were most frequent after the initial invasion of the fluids and were distinct from steady-state intermittent pathways by size and frequency.

The mobility of the nonwetting phase was observed to be different during transient flow. The change in saturation after percolation was small (the saturation was already 88% of the steady-state saturation at the time of percolation), but there was a much larger increase in pressure (the differential pressure was 70% of the steady-state differential pressure at percolation). This means that the change in saturation per unit time is smaller than the change in the total mobility of the fluids per unit time (which is inversely proportional to the pressure drop). The mobility of the nonwetting phase, for a given saturation, is higher for a lower differential pressure. This implies that the nonwetting phase was more mobile in the transitional stage than at steady state and that intermittency is associated with higher mobility of the nonwetting fluid phase. These results suggest that transient flow dynamics should not be modeled using steady-state flow properties. However, current models that do consider dynamic effects suggest these effects may be parameterized by rates of change of fluid saturation (Barenblatt et al., 2003; Hassanizadeh et al., 2002). This approach cannot be applied to our experiments because the saturation remains approximately constant while the total mobility changes significantly.

We estimate the length scale in the fluid plume front from which steady-state dynamics can be assumed and applied to scenarios, such as in Figure 1b. Steady state was achieved after the initial invasion in Experiment 1 after approximately 180 pore volumes of gas were injected through a sample 20 mm in length. This would correspond to a plume front governed by transient flow dynamics approximately 3.6 m thick. For Experiment 4, steady state was achieved after approximately 60 pore volumes of gas were injected, corresponding an intermittent event) whereas steady-state pathways involve much smaller events (always less than 5 pores) (Figure 8).

For experiments with higher amounts of intermittency at steady state (Figure 7), we would expect the transition to steady state to take longer. However, the experiments (2 and 3) with higher amounts of intermittency at steady state were done in sequence from Experiment 1. This suggests that the transition to steady state is history dependent. This is also evident in the pressure signal in Figure 6 as the oscillations persist, suggesting that the system wanted to retain its configuration, which would provide some resistance to the creation and destruction of pathways in other locations.
to a distance of approximately 0.9 m. This means the transition zone would be much smaller for systems such as conventional and heavy oil, and NAPL contamination, where intermittency is not expected at subsurface flow conditions. These numbers act as an order of magnitude estimate for the impact of transient dynamics on the characteristics of a plume. However, the transition zone would be heavily influenced by the lithology and would be site dependent.

There are added complexities in subsurface applications if the injected phase is already present, or if the plume is layered due to geological constraints. First, if the nonwetting phase is already present, then the transition to steady state is less evident than it was for the initial invasion of gas. Changing the flow parameters once the nonwetting phase had percolated the pore space resulted in gradual changes in saturation and intermittency as a new steady state was established. Second, for the Sleipner CO₂ plume, as an example, the plume exists as layers that are frequently 1–5 m thick (Chadwick et al., 2009). Our work suggests that the leading edge of the CO₂ plume in the unsteady-state regime is indeed more mobile than would be predicted by current quasi-steady-state models. This effect would be more noticeable in a layered plume with multiple plume fronts. These complexities may provide an explanation to why predictive modeling subsurface fluid flows at field relevant scales (100–1,000 m) is notoriously difficult, exemplified by the many attempts to model CO₂ plume migration in a saline aquifer at Sleipner (Cavanagh & Haszeldine, 2014; Chadwick & Noy, 2015; Singh et al., 2010; Williams & Chadwick, 2017; Zhu et al., 2015). Existing models that include transient dynamics focus on the rate dependence of saturation or capillary pressure, which do not necessarily change significantly in this transition (Barenblatt et al., 2003; Hassanizadeh et al., 2002; Hassanizadeh & Gray, 1990; Picchi & Battiato, 2018). Hence, we suggest that a new modeling framework should be explored to account for the behavior we have observed.

Data Availability Statement

The data for this paper are available on the Digital Rocks Portal (Spurin et al., 2020).

References


