Experimental analysis of spatial correlation effects on capillary trapping of supercritical CO$_2$ at the intermediate laboratory scale in heterogeneous porous media

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Abstract

Several numerical studies have demonstrated that the heterogeneous nature of typical sedimentary formations can favorably dampen the accumulation of mobile CO$_2$ phase underneath the caprock. Core flooding experiments have also shown that contrasts in capillary entry pressure can lead to buildup of non-wetting fluid phase (NWP) at interfaces between facies. Explicit representation of geological heterogeneity at the intermediate (cm-to-m) scale is a powerful approach to identify the key mechanisms that control multiphase flow dynamics in porous media. The ability to carefully control flow regime and permeability contrast at a scale that is relevant to CO$_2$ plume dynamics in saline formations offers valuable information to understand immiscible displacement processes and provides a benchmark for mathematical models. To provide insight into the impact of capillary heterogeneity on flow dynamics and trapping efficiency of supercritical CO$_2$ under successive drainage and imbibition conditions, we present an experimental investigation conducted in a synthetic sand reservoir. By mimicking the interplay of governing forces at reservoir conditions via application of surrogate fluids, we performed three immiscible displacement experiments to observe the entrapment of NWP in heterogeneous porous media. Capillary trapping performance is evaluated for each scenario through spatial and temporal variations of NWP saturation; for this reason we adopted x-ray attenuation to precisely measure phase saturations throughout the flow domain and apply spatial moment analysis. The sweeping performance of two different permeability fields with comparable variance but distinct spatial correlation was compared against a homogeneous base case with equivalent mean permeability by means of spatial moment analysis.
1. Introduction

Carbon Capture and Storage (CCS) is a technology that is under investigation to reduce atmospheric loading of CO\textsubscript{2} from power plants and heavy industries by injecting it into deep geological reservoirs, such as saline aquifers. In the storage formation, CO\textsubscript{2} will be in a supercritical state (scCO\textsubscript{2}) and its migration back to the surface will be prevented by the action of a set of physico-chemical mechanisms referred to as: (1) structural, (2) residual, (3) dissolution, and (4) mineral trapping [IPCC, 2005]. One of the key aspects that need to be considered is that these mechanisms affect each other in ways that are not readily apparent, since they occur over a wide range of time and length scales. Furthermore, the simultaneous occurrence of drainage and imbibition during the capillary-dominated post-injection stage has a direct effect on the spatial variation of the maximum injected-phase fluid saturation and consequently on the entrapped NWP saturation [Cihan et al., 2014]. Finally, the inherent heterogeneity of natural subsurface formations has a certain influence on the plume-reservoir interaction and on the effectiveness of each trapping mechanism. This complexity ultimately affects our ability to predict with confidence the fate of injected CO\textsubscript{2} and the success of the storage operation as a whole. The local capillary trapping phenomenon provides a good example of such complex behavior and it refers to the occurrence of locally high scCO\textsubscript{2} saturations (significantly above residual saturation) due to buildup of bulk phase behind so-called capillary barriers. The latter are associated with intra-reservoir heterogeneities of various sizes in the form of low-permeability features, such as lenses, lamina and/or beds [Ringrose et al., 1993]. On the
one hand, these barriers are considered beneficial to enhance the storage of CO\(_2\) for various reasons: their presence doesn’t depend on absolute seal integrity over an extended region, they can arrest the plume propagation both horizontally and vertically, and they allow for CO\(_2\) trapping at saturations larger than those expected from residual trapping alone [Green and Ennis-King, 2010; Hesse and Woods, 2010; Saadatpoor et al., 2010]. On the other hand, however, the presence of trapped CO\(_2\) significantly reduces the relative permeability to the resident brine, thus slowing down the mixing and subsequent dissolution process of the two phases.

As suggested by Koltermann and Gorelick [1996], the primary control on the flow paths of multiple fluids is exerted by hydraulic conductivity. With reference to the example described above, heterogeneities will therefore become relevant as soon as their presence significantly increases the overall variability in the permeability distribution of the given porous medium. Interestingly enough, permeability variations within length scales < 20 cm are reported for various depositional environments [Corbett et al., 1992]. Additionally, history dependent parameters of the capillary pressure-saturation curve, such as capillary entry pressure or residual NWP saturation, play a major role in controlling fluid displacement in a porous medium system governed by the interplay of viscous, buoyancy, and capillary forces, as it would be the case for scCO\(_2\)-brine flow in a geological formation. As a matter of fact, such a displacement is characterized by a variety of patterns influenced by heterogeneity and hysteresis, including lateral spreading, preferential flow and pooling [Held and Illangasekare, 1995; Illangasekare et al., 1995], as well as front pinning [Zhao et al., 2013] and blunting [Golding et al., 2013; Zhao et al., 2014], which in turns affect the overall sweep efficiency of the injection process. The ability to link spatial heterogeneity of key properties of multiphase porous media systems to the spreading and distribution of each fluid phase is needed for their full characterization.
So far, the phenomenon of capillary heterogeneity in CO₂ trapping has been studied experimentally, mostly by means of core-flooding experiments at the mm-cm scale [Krevor et al., 2011; Pini and Benson, 2013; Shi et al., 2011] and numerically, through reservoir- and core-scale simulations accounting for capillarity [Flett et al., 2007; Li and Benson, 2015; Saadatpoor et al., 2010]. While mathematical models need to be tested against experimental observations, laboratory investigations that use reservoir cores do not allow for spatial control of the distribution of the heterogeneity in two or three dimensions at the cm-m scale. In the latter case, despite recent advances in the application of imaging techniques, such as x-ray computed tomography, to the characterization of 3-D core-flooding experiments, the range of geometries that can be investigated is still limited by the nature and size of the core sample itself. In this context, the data sets from laboratory investigations of multiphase flow displacements in heterogeneous porous media at the intermediate (cm-meter) scale [Dawe et al., 2011; Hofstee et al., 1998; Kueper et al., 1989;] are rather scarce. Following the definition by Lenhard et al. [1995], the length scale of an intermediate-scale experiment has to be small enough for the laboratory setting to be controlled and the dimensions of the flow domain have to be compatible with measurement techniques. We add here that the usefulness of performing immiscible displacement experiments at the meter-scale lies in the ability to observe small-scale processes manifesting at a larger scale, allowing for quantification of their relative contributions to flow and transport phenomena. As pointed out by Oostrom et al. [2007] two-dimensional experiments conducted in intermediate-scale flow cells represent a suitable intermediary for mimicking flow processes occurring between one-dimensional column tests and field-scale experiments.

In a set of homogeneous experiments using unconsolidated sands in a 2-D sandpack of 70 cm by 16 cm [Trevisan et al., 2014], we have investigated the role of grain sizes on the influence
of capillary forces on the displacement of a non-wetting phase (NWP) plume. Fine-grained sands increase the relative contribution of capillary forces vs. gravity and viscous forces, thus stabilizing the front and leading to larger footprints and lower average NWP saturations. Furthermore, the potential mobilization of temporarily trapped NWP at elevated saturations was demonstrated with the application of a forced wetting phase flow once the fluids had reached a complete redistribution. The addition of heterogeneous structures to those fairly simple experiments would allow extending such investigations to the study of capillary barrier phenomena on NWP plume migration and trapping. In this context, a key aspect to take into account is how such heterogeneities are correlated [Bryant et al., 2008; Han et al., 2010; Ide et al., 2007]. In the present study we consider two heterogeneous scenarios characterized by different continuity of the sand zones and we compare them with a homogeneous base case conducted previously, so as to observe and quantify the effects of sand correlation on the lateral migration and entrapment of a NWP plume. As expected, correlated heterogeneities led to more pronounced horizontal propagation of the plume, whereas uncorrelated heterogeneities led to wider range of NWP saturations and larger amount of trapped fraction. Although the synthetic cases presented in this study are not directly scalable to real storage scenarios and the well-defined heterogeneities do not resemble any realistic sedimentary structure, such experiments enable direct insight of the contribution of different multiphase transport phenomena at the scale and flow regime that are relevant to CO₂ plume dynamics in saline formations. In the context of spatial correlation of capillary heterogeneity, this study focuses on the influence of local capillary barriers at the cm-to-m scale with the purpose of providing a dataset for verification of mathematical models.
2. Experimental approach and methods

The immiscible displacement experiments were carried out in a quasi-two-dimensional sand pack enclosed by the same rectangular flow-cell with internal dimensions \((L \times W \times H)\) of \((91.4 \times 5.6 \times 61)\) cm\(^3\) described in a previous paper (Trevisan et al. [2014], see Figure 1). The transparent acrylic walls allow for visual inspection of the displacement process, so that digital photographs and saturation distribution maps gathered via x-ray attenuation can be directly compared. The flow domain consists of a gently sloping \((2^\circ)\) aquifer with an aspect ratio \((L/H)\) of 4.375 and initially saturated with the wetting phase. Details regarding the preparation of the synthetic aquifer and control of the boundary conditions, together with the procedure for the acquisition of the 2-D x-ray images are reported in Trevisan et al. [2014].

![Figure 1](image_url)  
**Figure 1** Synthetic aquifer used for the experiments presented in this study. Injection is carried out from the left by applying a constant head \((\Delta H)\) over the entire cross-section of the aquifer (drainage and forced imbibition); during fluid redistribution no injection takes place. The aquifer has a gentle slope \((2^\circ)\) in the direction of flow.
In the present study, two heterogeneous sand configurations (Het.1 and Het.2) with different spatial correlation of the permeability field (Figure 2) are considered. In both cases, the background material is the same sand used in the homogeneous experiment with intermediate permeability, while finer and coarser sands are distributed within the domain by following two correlation values. Each experiment involves three stages, namely non-wetting fluid injection or drainage (5.5 hours), fluid redistribution (no injection for approximately two weeks), and forced imbibition stages (48 hours). Experimental parameters for the drainage and forced imbibition stages are given in Table 1.

Table 1 Summary of the experimental parameters. Ca is capillary number \((U_i\mu_i/\sigma)\), where \(U_i\) and \(\mu_i\) are the Darcy velocity and viscosity of the injected fluid, respectively, and \(\sigma\) is the interfacial tension between the injected and displaced fluids; \(\Delta H\) is the height difference between constant pressure reservoir and constant head outflow boundary; PV is the pore volume of each aquifer measured with x-rays; PVI is the pore volume injected during each stage.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>stage</th>
<th>flow rate (ml/min)</th>
<th>time (h)</th>
<th>Ca (-)</th>
<th>(\Delta H) (cm)</th>
<th>PV (cm(^3))</th>
<th>PVI (-)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hom.</td>
<td>Drainage</td>
<td>0.7</td>
<td>5.5</td>
<td>(1.0\times10^{-5})</td>
<td>35</td>
<td>2379</td>
<td>0.097</td>
</tr>
<tr>
<td></td>
<td>F.I.</td>
<td>2</td>
<td>48</td>
<td>(3.6\times10^{-4})</td>
<td>19</td>
<td></td>
<td>2.42</td>
</tr>
<tr>
<td>Het. 1</td>
<td>Drainage</td>
<td>0.72</td>
<td>5.5</td>
<td>(1.0\times10^{-5})</td>
<td>35</td>
<td>0.109</td>
<td></td>
</tr>
<tr>
<td></td>
<td>F.I. I</td>
<td>1</td>
<td>24</td>
<td>(1.8\times10^{-4})</td>
<td>13</td>
<td>2164</td>
<td>0.665</td>
</tr>
<tr>
<td></td>
<td>F.I. II</td>
<td>1.5</td>
<td>24</td>
<td>(2.7\times10^{-4})</td>
<td>17</td>
<td></td>
<td>0.998</td>
</tr>
<tr>
<td></td>
<td>Drainage</td>
<td>0.72</td>
<td>5.5</td>
<td>(1.0\times10^{-5})</td>
<td>35</td>
<td>0.106</td>
<td></td>
</tr>
<tr>
<td>Het. 2</td>
<td>F.I. I</td>
<td>1</td>
<td>24</td>
<td>(1.8\times10^{-4})</td>
<td>13</td>
<td>2238</td>
<td>0.643</td>
</tr>
<tr>
<td></td>
<td>F.I. II</td>
<td>1.5</td>
<td>24</td>
<td>(2.7\times10^{-4})</td>
<td>17</td>
<td></td>
<td>0.965</td>
</tr>
</tbody>
</table>

In addition to visual inspection of the aquifer by means of x-rays, spreading of the plume throughout the various stages of the experiments is quantitatively assessed by following a common approach that is based on the analysis of first spatial moments [Eichel et al., 2005;
Fagerlund et al., 2007; Han et al., 2010; Kueper and Frind, 1991]. For an immiscible plume, the moments are defined in a similar manner as for a solute concentration [Freyberg, 1986]:

\[
M_j(t) = \iint \varphi(x,z)S_{NW}(x,z,t)x^jz^j \, dx \, dz
\] (1)

Where \( \varphi \) is the porosity (space-dependent), \( S_{NW} \) is the NWP saturation (space- and time-dependent), and \( x, z \) are the horizontal and vertical spatial coordinates, respectively. Since the measured saturations are averaged across the width of the flow-cell, the \( y \)-direction is neglected in the calculation of the spatial moments. The first moments in the \( x \)- and \( z \)-direction normalized by the total NWP volume present in the domain (\( M_{00} \)) describe the coordinates of the center of mass of the plume:

\[
x_c = \frac{M_{10}}{M_{00}}
\] (2)

\[
z_c = \frac{M_{01}}{M_{00}}
\] (3)

In order to evaluate trapping effectiveness for the heterogeneous experiments, we compare the observed initial-residual NWP saturations with the Land trapping model [Land, 1968], which takes the following form:

\[
S^*_{nw,r} = \frac{S^*_{nw,i}}{1 + CS^*_{nw,i}}
\] (4)

where \( S^*_{nw,r} \) is the effective residual NWP saturation, \( S^*_{nw,i} \) is the effective maximum (or initial) NWP saturation reached at flow reversal, and \( C \) is the land trapping coefficient, which is a function of the effective endpoint residual NWP saturation, \( S^*_{nw,r} \) [Land, 1968]:

\[
C = \frac{1}{S^*_{nw,r}} - 1
\] (5)

All effective saturations are calculated by normalizing measured NWP saturations by the volume of wetting phase contributing to the flow:
\[ S_{\text{nw}}^* = \frac{S_{\text{nw}}}{1 - S_{\text{w,irr}}} \]  \hspace{1cm} (6)

where \( S_{\text{w,irr}} \) is the irreducible wetting phase saturation, which takes a value of 0.07 for all the three sands.

### 2.1 Porous media

Three hydrophilic silica sands (Accusand, Unimin corp.) with effective sieve numbers #30/40, #40/50, and #50/70 were used for packing the synthetic aquifers. By using these grades, the flow domain was populated with three levels of permeability ranging from \(3.5 \times 10^{-11} \) to \(1.1 \times 10^{-10} \text{ m}^2\). The main physical properties of the sands used in this work have been characterized previously [Yoon et al., 2008] and are reported in Table 2. Both porosity \( (\varphi_{\text{avg}}) \) and uniformity coefficient \( (d_{60}/d_{10}) \) are very similar for coarse and medium sands, while the finer sand shows slightly larger values. Moreover, although the differences in capillary entry pressure and endpoint values (residual NWP saturation, \( S_{\text{nw,r}}^{*\text{max}} \), and irreducible wetting phase saturation, \( S_{\text{w,irr}} \)) between the three sands are minimal (~0.1 kPa), we expect that at sufficiently low flow rates, the effect of the different properties of the sands will affect the displacement and, accordingly, the saturation distribution.

<table>
<thead>
<tr>
<th>Material</th>
<th>Sieve size</th>
<th>( k (\text{m}^2) )</th>
<th>( \ln k )</th>
<th>( \varphi_{\text{avg}} )</th>
<th>( d_{50} ) (mm)</th>
<th>( d_{60}/d_{10} ) (-)</th>
<th>( S_{\text{nw,r}}^{*\text{max}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 (coarse)</td>
<td>#30/40</td>
<td>(1.14 \times 10^{-10} )</td>
<td>-22.89</td>
<td>0.355(^a)</td>
<td>0.53(^a)</td>
<td>1.22(^b)</td>
<td>0.22</td>
</tr>
<tr>
<td>2 (medium)</td>
<td>#40/50</td>
<td>(6.43 \times 10^{-11} )</td>
<td>-23.47</td>
<td>0.355(^a)</td>
<td>0.36(^a)</td>
<td>1.21(^b)</td>
<td>0.19</td>
</tr>
<tr>
<td>3 (fine)</td>
<td>#50/70</td>
<td>(3.46 \times 10^{-11} )</td>
<td>-24.09</td>
<td>0.372(^a)</td>
<td>0.26(^a)</td>
<td>1.28(^b)</td>
<td>0.18</td>
</tr>
</tbody>
</table>

\(^a\) from Yoon et al. [2008].  
\(^b\) estimated from sieve data provided by the manufacturer.
The two heterogeneous permeability fields have the same volumetric proportions of the three sands (15%, 66%, and 19% for coarse, medium, and fine sand, respectively) and were generated to create two scenarios with different spatial correlations (Figure 2), namely poorly correlated (Het. 1, also referred to as “uncorrelated”) and moderately correlated (Het. 2, also referred to as “correlated”). The packing grid consists of 224 cells (28 columns by 8 rows) and each grid block has dimensions of (2.5 x 2) cm$^2$.

**Figure 2** Spatial distribution of sand blocks with variable permeability; #30/40 (no. 1) is colored in yellow, #40/50 (no. 2) in green, and #50/70 (no. 3) in blue. Experimental variograms corresponding to the two scenarios are presented in

**Figure 3.**
Figure 3 shows the experimental variograms along the longitudinal flow direction in terms of a semivariogram function $\gamma(h)$ plotted as a function of a lag distance $h$ for both scenarios. In the first case (Het. 1) the maximum variability value (sill) is reached at very short distances, thus indicating poor spatial correlation, and the progression is scattered around an average value of 0.38, thus suggesting a cyclic behavior. On the contrary, the semivariogram of the second case (Het. 2) increases with distance and approaches a value of 0.38 at a distance equal to 25 cm (range). It is worth noting that the variance of both permeability fields is the same (0.38) and agrees with the variance of the normal distribution function computed on a random distribution of the three sands in the volumetric proportions used in this study.
Figure 3 (left) $P_c$-S constitutive relationships for the three sands used in the experiments. Van Genuchten model is fitted to experimental data measured by Mori et al. [2015a]. Continuous and broken lines represent primary drainage and main wetting cycles, respectively; (right) experimental variograms along x-direction of the two heterogeneous cases. The vertical dashed line indicates the distance at which data are no longer autocorrelated (range) for Het. 2. The horizontal dashed line shows the variance (sill) shared by both heterogeneous scenarios.

### 2.2 Fluids

In order to avoid experimental complexity associated to high-pressure flow-cells at the meter-scale, and to achieve displacement velocities compatible with the measurement time of the x-ray attenuation device, the experiments were conducted with a pair of surrogate fluids at ambient conditions. Fluids have been identified that mimic the density and viscosity contrasts of scCO$_2$ and brine at reservoir $P,T$ conditions as reported in Table 3. In particular, we selected an isoparaffinic oil (Soltrol 220) and a glycerol-water mixture (80:20 w/w) to represent the injected (non-wetting) and displaced (wetting) phases, respectively. In order to allow for direct visualization of the flow and to increase x-ray attenuation contrast with the wetting phase, Soltrol 220 was dyed red with Sudan IV (Fisher Scientific) and doped with 10% w/w Iodoheptane (Alfa Aesar).

<table>
<thead>
<tr>
<th>phase</th>
<th>$\rho$ (kg/m$^3$)</th>
<th>$\mu$ (mPa·s)</th>
<th>$\mu_{nw}/\mu_w$</th>
<th>$\rho_{nw}/\rho_w$</th>
<th>IFT (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Soltrol 220</td>
<td>860 (1210)</td>
<td>4.9 (61)</td>
<td>0.072</td>
<td>0.71</td>
<td>15</td>
</tr>
<tr>
<td>Glycerol-water</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>scCO$_2$</td>
<td>266-733$^a$ (760$^c$)</td>
<td>0.023-0.0611$^a$ (0.06$^c$)</td>
<td>0.026-0.20$^a$ (0.075$^c$)</td>
<td>0.22-0.75$^a$ (0.745$^c$)</td>
<td>19.8$^b$</td>
</tr>
<tr>
<td>Brine</td>
<td>945-1230$^a$ (1020$^c$)</td>
<td>0.195-1.58$^a$ (0.8$^c$)</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^a$ estimates from Nordbotten et al. [2005], $T = 35-155^\circ$C, $P = 10.5-31.5$ MPa

$^b$ measurement from Bennion and Bachu [2006], $T = 43^\circ$C, $P = 20$ MPa, brine salinity = 2.7% wt.
estimates from Singh et al. [2010] for Sleipner field

3. Experimental results

In order to identify the displacement and trapping mechanisms associated with drainage and forced imbibition, we describe these two stages separately in the following sections.

3.1 Drainage and fluid redistribution

Digital photographs of the injected plume provide an effective method to track plume movement over time and to qualitatively detect distinctive displacement patterns among the different packing configurations (Figure 4 and Figure S1). As an example of general validity, Figure 4 shows for the three scenarios snapshots of the plume taken just after injection has ceased and at the end of the fluid redistribution period (about 2 weeks), i.e., prior to starting the forced imbibition campaign. In the former case (left column), it can be readily observed that the plume has advanced less for the uncorrelated permeability field (Het. 1) as compared to the other two cases, where its leading edge has already reached about ¾ of the total longitudinal extent of the aquifer. For the heterogeneous scenarios, the spreading of the plumes is highly controlled by the arrangement of low- and high-permeability zones that characterize these systems: in the case of poorly correlated heterogeneity (Het. 1), a more compact plume is observed, in contrast to a channeling behavior exhibited in the spatially correlated heterogeneous experiment (Het. 2). As expected, during those experiments only the coarse and medium permeability sands were invaded by the NWP, since the pressure drop driving the NWP injection during the heterogeneous experiments was kept at the exact same constant level.

Figure 4 (right) shows that once the injection has ceased, the NWP plumes continue to redistribute, mainly due to buoyancy and capillary forces. Here, the striking difference among the three experiments is that NWP breakthrough at the outflow boundary was observed for both
the homogenous and the spatially correlated experiment, while the plume movement stopped and remained entirely trapped in the aquifer for the uncorrelated experiment. Moreover, the intensity of the colors in those snapshots further suggests that in this latter case (Het. 1) the NWP is trapped at relatively large saturations as compared to the other two scenarios. We anticipate that this phenomenon may have important implications in the potential remobilization of the plume.

Figure 4 Digital photographs showing plume distribution for homogeneous, heterogeneous uncorrelated, and heterogeneous correlated configurations after injection (left) and fluid redistribution (right) stages (scale bar in mm).

A more quantitative assessment on the evolutions just described is achieved by comparing the coordinates of the center of mass of the plume for the three experiments, as given by Eqs. (1)-(3) and shown in Figure 5. In particular, the temporal evolution of the center of mass along the x-direction (Figure 5, left) suggests that (1) the spatially uncorrelated (Het. 1) and the homogeneous (Hom.) experiments exhibit a common displacement speed during the fluid redistribution period, and that (2) the plume migrates significantly faster through the spatially
correlated heterogeneous aquifer (Het. 2). Note that the former observation does not contradict
the conclusions drawn from the analysis of the photo images shown in Figure 4, but rather hints
towards a different aspect ratio of the plume. In other words, the extended tongue that
characterizes the homogeneous plume carries less weight into the estimation of the center of
mass in the horizontal direction. The analysis of the vertical position of the plume’s center of
mass (Figure 5, right) supports this conclusion with the height of the center of mass decreasing in
the direction Hom. > Het. 2 > Het. 1. Generally, a fairly stable evolution is observed in each
experiment, with the total variation of the height of the center of mass over time being less than 1
cm. Interestingly, a common feature shared by experiments Hom. and Het. 2 is the downward
movement of the center of mass as the plume breaks through the outflow boundary (filled
symbols in Figure 5). This could be due to the depletion of NWP from the upper region of the
plume, leading to an apparent downward movement of the center of mass. This behavior is not
observed in Het. 1 (the slower and more compact plume), where the vertical position of the
center of mass has stabilized at about 200 hours. The fluid redistribution stage was considered to
be over when the center of mass reached equilibrium and no further movement occurred along
horizontal and vertical directions. This period lasted slightly less than two weeks for both
homogeneous and heterogeneous experiments.

![Graphs showing changes in center of mass over time for different experiments.](image-url)
Figure 5 Temporal evolution of the horizontal (left) and vertical (right) coordinates of the plume’s center of mass. Filled symbols represent measurements taken after plume breakthrough into outflow boundary. Horizontal coordinate of well’s center is -750 mm. Data is available in the supplementary information.

3.2 Forced imbibition

At the conclusion of the fluid redistribution period, a flow of glycerol-water (the wetting phase) was imposed throughout the aquifer, thus mimicking a “chase-brine” type of event [Juanes et al., 2006; Qi et al., 2009; Rahunanthan et al., 2014]. This forced imbibition consisted of two consecutive 24-hour periods of different flow rates to trigger the mobilization of NWP that has accumulated behind capillary barriers. The first stage was carried out at 1 ml/min ($Q_1$), i.e. injecting 0.67 pore volumes, and it was followed by the injection of 1 pore volume of wetting phase at 1.5 ml/min ($Q_2$). For the sake of completeness, we include in the discussion results obtained for the homogeneous experiment, where the forced imbibition was carried out in one stage and by applying a constant flow rate of 2 ml/min. These flow-rates correspond to capillary numbers of $1.8 \times 10^{-4}$ (1 ml/min), $2.7 \times 10^{-4}$ (1.5 ml/min), and $3.6 \times 10^{-4}$ (2 ml/min), representing a transition zone between capillary- and viscous-dominated flow regimes, which could lead to difficulties in predicting experimental results with conventional continuum-based multiphase flow models.

Figure 6 provides an overview of the three experiments in terms of box plots that illustrate the range of NWP saturations measured before and after forced imbibition events from the x-ray scanning; larger boxes, enclosed by 1st and 3rd quartiles, represent plumes with a wide range of NWP saturations, in contrast with narrow boxes denoting plume saturations constrained to a limited range of values. In the figures, the dashed horizontal lines represent the endpoint residual NWP saturation for the background sand, $S_{max}^{nw}$, as obtained from final NWP saturation
measurements in the homogenous setup [Trevisan et al., 2014]. In other words, any value of residual saturation measured below this line can in principle be described by a trapping model, such as the one proposed by Land [1968] introduced in Section 2.

Figure 6 Box plots showing NWP saturation distribution before and after forced imbibition (F.I.) events. Middle lines represent the median of the distributions, upper and lower edges represent first and third quartile, respectively, while ‘whiskers’ above and below each box represent maximum and minimum values observed, respectively. Dashed line represents the endpoint residual NWP saturation of #40/50 sand.

Several observations can be made from these figures. In the homogeneous scenario (left panel), most of the plume is found at saturations below the maximum residual saturation limit already after the fluids’ redistribution period. Accordingly, a forced imbibition event has the effect of further reducing the NWP saturations, thus leading to a situation where the entire plume is trapped with a very narrow distribution of saturations around a mean value of 0.08. On the contrary, distinct responses to the forced imbibition are observed when the two heterogeneous configurations are considered. For the Het. 1 case (center), most of the plume still contains saturations significantly larger than $S_{nw,max}$ after the first forced imbibition event, with about 20% of the distribution found within a range of saturation values 0.4 - 0.8. As expected, the second event performed at a larger flow rate is more effective and leads to a narrower range of saturation...
values that are mostly found below and just above the residual limit, $S_{\text{nw, t}}^\text{max}$. Interestingly, a similar final distribution is observed for the Het. 2 case (right panel), although in this case the latter was attained already after the first event. Het. 2 shows a similar spread of saturation values prior to the wetting phase injection as for the homogeneous case. In both heterogeneous cases, however, the presence of NWP saturation higher than residual, represented by the top ‘whiskers’, is evidence of capillary barrier effect still holding part of the plume in a continuous phase. As described below, more insights on this phenomenon are obtained by considering the spatial distribution of the plume at various stages of the process.

2-D plume saturations distributions measured via x-ray attenuation after these forced imbibition events are shown in Figure 7 and are compared to images taken just prior to the injection of the wetting phase (a more comprehensive set of images is provided in Figure S2).

![Figure 7 Comparison of 2-D NWP saturation distribution measured via x-ray attenuation for homogeneous (top), spatially uncorrelated (middle row), and correlated (bottom row) heterogeneous experiments prior and subsequently to forced imbibition events at different flow rates.](image)

It appears that, in all three cases the forced imbibition has indeed mobilized the plume, though with different effectiveness. Although the injection of wetting phase into the homogenous
aquifer did not result in an effective mobilization of the plume, it can be seen that the region of
trapped NWP left behind is fairly homogeneous and has maximum non-wetting phase residual
saturation levels that would be expected from the residual saturation captured in the capillary
pressure curve alone (about 0.15). This region is concentrated near the injection well, this being
the same region where most NWP was present prior to later injection of the wetting phase, as a
result of the interplay between viscous and buoyant forces in a sloping aquifer. This picture is
very different when the heterogeneous scenarios are considered. After the first imbibition event
and in both cases, the plume occupies almost entirely the top half of the aquifer; however, while
for Het. 1 trapped saturations are locally still very high (up to 0.80), most of the plume in Het. 2
contains NWP at saturations close to the residual limit observed for the homogeneous case. In
this context, therefore, capillary forces help distributing the plume more homogeneously
throughout the entire aquifer, thus counteracting the effect of buoyancy; at the same time, the
spatial distribution of potential capillary barriers is key in controlling the actual saturation of the
trapped plume, this being very different when Het. 1 and Het. 2 are compared. In fact, once the
bulk fraction of the plumes has been swept by the second (and stronger) forced imbibition event,
the plume in both heterogeneous cases are practically indistinguishable, these having very
similar footprint and saturation distribution. This outcome is amenable to the increased
magnitude of viscous forces over the retention capability of capillary forces and offers a direct
insight into the correlation between flow regimes and residual saturation, also illustrated by the
capillary desaturation curve [see, for example, Larson et al., 1981]. It is worth noting that despite
the high NWP saturations, and therefore high NWP relative permeability, occurring after the first
forced imbibition event in the Het.1 case, the plume advancement is still significantly hindered
by the several capillary barriers; this behavior cannot be captured if relative permeability alone is considered as an indicator of fluid mobility in the aquifer.

4. Discussion

The present study consists of an experimentally driven analysis on migration and entrapment under capillary-dominated flow conditions of a scCO$_2$/brine-surrogate fluid pair in an unconsolidated sand reservoir that is embedded with structurally well-defined heterogeneities. The characteristic length of the heterogeneities considered in this study (from about ($2.5 \times 2$) cm$^2$ to ($15 \times 2$) cm$^2$) is of the same order in size as the “synthetic” aquifer, that is ($16 \times 70$) cm$^2$, and can therefore be associated with bedform type of heterogeneities encountered in sedimentary formations [Ringrose et al., 1993]. These heterogeneities are implemented in the form of either barriers or high-permeability streaks and their regular geometry allows for a systematic investigation on the contribution of capillary phenomena to the trapping and migration of the NWP. Two scenarios were presented that differ in the spatial configuration (or correlation) of the heterogeneities and a comparison has been made with results from an immiscible displacement experiment performed in a homogeneous aquifer (base-case). X-ray imaging has been applied to quantify the temporal and spatial distribution of fluids’ saturations throughout the various stages of the experiments, which in each case included a period of injection, fluids’ redistribution and forced imbibition. The latter represents a “chase-brine” type of event that is often considered in the design of field-scale injection operations in saline aquifer as a strategy to maximize CO$_2$ storage, both through capillary trapping and dissolution [Hassanzadeh et al., 2009; Qi et al., 2009; Saeedi et al., 2011].
4.1 Displacement and trapping mechanisms

As shown in Figure 8 for the Het. 1 configuration, a more detailed analysis of the mechanisms associated with capillary barriers and forced imbibition driving the trapping and mobilization of NWP can be carried out by observing the evolution of the NWP saturation at selected locations. Generally, the presence of heterogeneities leads to abrupt variations in NWP saturation at contiguous locations, and depending on the relative position of the capillary barrier (upstream vs. downstream and distance from the injection well) distinct trends in the temporal evolution of the saturation are observed. In particular, the following mechanisms have been identified as a response to a forced imbibition event: (1) invasion of NWP into previously inaccessible sands (e.g. points A, D, and E); (2) increase in NWP saturation (e.g. all points in (1) with addition of F); (3) ‘wave’ effect reflecting the transit of mobile NWP characterized by the attainment of a maximum NWP saturation followed by a decrease; the magnitude of this ‘wave’ increases at locations farther from the injection well, facilitated by larger volumes of NWP accumulating at near-well regions; (4) rapid decrease of NWP saturation as a consequence of wetting phase flow mobilizing the accumulated bulk NWP from coarse sands (e.g. point B). It is worth noting that the uncorrelated nature of the distribution of heterogeneities in the Het.1 experiment has the effect of increasing the frequency of these mechanisms. Most importantly, not only a forced imbibition event results in the immobilization of NWP into a residual phase (as it would also happen in a homogeneous aquifer), but it leads to a local increase of NWP saturation and in some instances it can even force NWP into zones of the aquifer previously not accessible. The latter observation suggests that room exists for optimizing CO₂ storage through a combination of NWP and wetting phase injection into the aquifer. The mechanisms mentioned
above are responsible for the different NWP trapping performance exhibited by the three configurations, as reported in Table 4.

![Image: NWP saturation evolution observed at six selected locations in the uncorrelated heterogeneous scenario (Het. 1). Vertical dashed lines indicate the onset of the forced imbibition phase. Data is available in the supplementary information.]

### 4.2 Trapping performance

Analysis of total NWP volume retained in the aquifer provided by zero-th moment calculation was compared with a simple material balance calculation (NWP injected versus NWP collected at outflow) to give a range of the trapping performance for each experiment throughout the three stages described above (an overview is given in Table 4). As anticipated in the results section, residual NWP saturations (above 0.16 for these sands) could already be attained within most part of the plume after the fluids’ redistribution period (no injection) in the homogeneous case (43 vol.% of the injected volume retained in the aquifer); accordingly, a weak effect of forced imbibition on saturation distribution was observed and about 19 vol.% of the plume was...
mobilized and produced. While the implementation of correlated heterogeneities (Het. 2) enhanced the retention of the plume behind capillary barriers (with NWP saturations up to 0.5 and a trapping of 68 vol.%), once the first forced imbibition was put in place 43 vol.% of the plume’s volume was remobilized and a total of 55 vol.% has left the aquifer after completion of the second stage (similarly to the homogeneous case, 37 vol.% of the injected volume is retained in the aquifer). The uncorrelated heterogeneity in Het. 1 represents the most desirable arrangement of capillary barriers, since the plume was retained entirely in the aquifer before the onset of forced imbibition and only 6 vol.% of its volume was mobilized and produced after the first stage of wetting phase injection. After completion of the second stage, however, the final trapped NWP volume showed a value similar to the other two experiments, with 42 % of the plume’s volume being trapped.

<table>
<thead>
<tr>
<th></th>
<th>Hom.*</th>
<th>Het. 1</th>
<th>Het. 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>Trapped NWP fraction before onset of forced imbibition</td>
<td>39-47%</td>
<td>100%</td>
<td>64-72%</td>
</tr>
<tr>
<td>Trapped NWP fraction after 1st forced imbibition</td>
<td>N/A</td>
<td>90-98%</td>
<td>35-43%</td>
</tr>
<tr>
<td>Trapped NWP fraction after 2nd forced imbibition</td>
<td>31-39%</td>
<td>38-46%</td>
<td>33-41%</td>
</tr>
</tbody>
</table>

8% range is a result of the NWP volume around the injection well that is not detectable by x-ray attenuation.

N/A: not applicable.

Only one forced imbibition event performed.

In a so-called ‘Initial-Residual’ saturation plots (Figure 9), the effective residual saturation observed after a waterflood ($S_{nw,i}^*$) is plotted as a function of the corresponding effective maximum NWP saturation reached at flow reversal ($S_{nw,j}^*$). This type of plot enables the development of several parametric models that describe this correlation [Jerauld, 1997; Land,
known under the general name of “trapping models”. When applied to the experiments presented in this study, such plot can be quite useful as it can be populated with saturation values measured at different spatial locations before and after a forced imbibition event, thus including the possibility to distinguish among different types of sands.

Figure 9 provides a comparison between results from the two heterogeneous cases (shown as symbols for both coarse (#30/40) and medium (#40/50) sands) together with predictions of the Land trapping model (shown as solid lines) applied to results from two homogeneous scenarios using the same sands and that were published in a previous paper. As reported in a previous publication [Trevisan et al., 2014], these curves were generated from equation 4, while Land’s coefficients for the two sands were calculated from equation 5 by using the corresponding effective endpoint residual saturation, \( S_{\text{nw},r}^* \). For the Het.1 case and prior to the forced imbibition event, all saturation points lie close to the identity (1:1) line that represents a scenario with essentially no mobilization (\( C=0 \) in Equation 4). For coarser sands this effect is the strongest due to the higher probability for the latter of being located behind a sand with higher entry pressure. A similar pattern can be observed for the Het. 2 case, where both sands lie now underneath the identity line, but with the finer sand showing significant mobilization already during the fluids’ redistribution stage. The stronger trapping effectiveness seen in the Het. 1 case as compared to the Het. 2 case is expected due to the higher occurrence of capillary barriers. Interestingly, when the situations are compared after the forced imbibition events, residual saturations have significantly dropped and occupy a region bounded by the Land’s trapping model curves that represent coarse and medium sands. This is clearly evident in the Het. 2 case, while trapped saturation values above residual are still present at some locations within
the coarser sand zones of the Het. 1 case. This behavior supports the convenience of Land’s trapping model for predicting residual NWP saturations as a function of maximum initial NWP saturations also in heterogeneous scenarios, when a forced wetting phase flow has swept the bulk of NWP accumulating at above-residual saturations behind capillary barriers. This observation is indeed very important as it suggests that a heterogeneous system can eventually be treated as homogeneous when the ultimate (long-term) trapping behavior is considered. It is worth noting, however, that although the experimental points lie on the same curves, the ranges of saturations among the different scenarios are quite different, with the cloud of points moving towards the left-hand side of the plot in the direction Het.1 < Het. 2 < Hom. This trend reflects the fact that much larger local saturation values can be reached in the presence of heterogeneities, as pointed out in the Results section. Moreover, those saturations have a stronger potential of being mobilized when the system is subject to forced imbibition, as a larger saturation implies a larger relative permeability.

As a final remark, it is worthwhile to analyze these results in the context of large-scale field’s operation. As anticipated above, the forced imbibition applied in this study can be related to a “chase brine” scheme that is adopted to enhance the displacement efficiency and trapping capacity in a reservoir [Juanes et al., 2006; Qi et al., 2009]. Accordingly, we can ask ourselves when the mobilization of part of the plume’s volume trapped behind capillary barriers will eventually stabilize into a Land-type behavior. For a simplified scenario of a 1-D reservoir, such estimate can be readily obtained as:

\[ t = \frac{PV}{Q} = \frac{L \cdot \phi}{u} \]  

(7)

where \( L \) is the longitudinal extension of the reservoir, \( \phi \) is its porosity and \( u \) is the Darcy velocity of the waterflood. The latter is often assumed to take a value of about 0.3 m/day
[Ringrose et al., 1993]; for a reservoir with a 10-50 km longitudinal extension and an arbitrary porosity of 0.2 it would take 20 to 100 years to reach injection of 1 Pore Volume, i.e. the forced imbibition scheme adopted in this experimental study.

![Figure 9](image)

**Figure 9** Trapped NWP saturations measured in the coarse (triangles) and medium (circles) sands before (empty symbols) and after (filled symbols) forced imbibition as a function of the maximum NWP saturation reached at flow reversal. The dashed line shows the 100% trapping (Land coefficient $C=0$), while blue and red lines represent Land trapping models derived from endpoint residual NWP saturations observed in $P_c$-$S$ relationships of coarse and medium sands, respectively.

Data is available in the supplementary information.

5. **Concluding remarks**

In order to test the predictive ability of detailed mathematical simulations of fluid transport in geological media, experimental studies are needed that quantitatively characterize (1) the spatial heterogeneity of geological media, (2) the distribution of fluid phases in terms of saturation and spatial extents, (3) the temporal evolution of the plume at various stages of the process. By carrying out the intermediate scale experiments presented in this investigation we
intend to create a unique data set that highlights the usefulness of such information to understand
the influence of heterogeneity correlation on fluid migration and trapping capacity. Although we
are aware of the significant role played by small-scale heterogeneity in governing multiphase
flow through reservoir rocks, this study has focused on the effects of larger scale features that
eventually will also contribute to the final distribution of a scCO$_2$ plume in a brine-saturated
reservoir. Two well-defined arrangements of three sands are used to represent a spatial correlated
and an uncorrelated scenario to be compared to the medium permeability homogeneous scenario
presented in chapter 4. From the observation of the experimental results presented in this
investigation, the following conclusions are drawn:

1. Spatial variability of capillary entry pressure exerts a major control on NWP plumes
   propagating in a capillary-dominated flow regime. Neglecting this variation will lead to a
   misrepresentation of plume’s configuration as well as underestimation of NWP saturation
   trapped behind capillary barriers and overestimation of plume’s front advancement;

2. Spatial correlation of sand bodies influences the ability of capillary barriers to prevent
   migration of NWP plume. Characterization of variance alone does not allow for prediction of
   capillary barrier performance, unless flow is occurring in a viscous-dominated regime;

3. Under capillary-dominated flow conditions, the ability of capillary barriers to hinder plume
   advancement cannot be captured if relative permeability alone is considered as an indicator
   of fluid mobility in the aquifer;

4. Heterogeneity influences plume spreading mainly during the short-term injection period,
   while long-term trapping performance of heterogeneous models can be reproduced by Land’s
   models fitted to homogeneous models with analogous mean permeability;
5. The average behavior of both homogeneous and uncorrelated heterogeneous models are similar, as shown by first moment analysis, although spatial variation of saturation values show significant differences between them. This last remark implies that (1) experiments that consider heterogeneities are needed to make observations on the extent of local saturation variations, (2) the non-wetting phase may be re-mobilized at locations where saturation is high and (3) a very careful treatment of the $P_c$-$k_r$-$S$ relationship would be required in a simulator to capture this behavior.

As a part of the continuing research effort, the presented study will be extended to include a greater degree of heterogeneity as well as larger domain size that will allow for larger contribution of buoyancy forces to the plume displacement. Larger setups will also allow to study the influence of background hydraulic gradient and sequential injection methods on plume migration and trapping. Furthermore, continuum-based numerical simulations will be carried out to test the significance of capillary heterogeneity, following the work of Mori et al. [2015b].

6. Acknowledgements

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