Chapter 4

GRAVITY DESTABILISED GAS FLOW REGIMES

4.1 Introduction

The pore-scale mechanisms governing gravity-driven regime transitions during a depressurization process have been investigated by Ezeuko et al. (2010) and McDougall and Mackay (1998). An analogous study on gas injection has, however, not been published to date and the pore-scale mechanisms involved are yet to be fully explored. This chapter sets out to address this by presenting analyses of flow regimes associated with gas injection through liquid-saturated porous media. A flow regime classification scheme is presented which describes the gas saturation distribution patterns that emerge in response to variations in fluid and rock (network) properties, including: the mean pore (capillary) radius, interfacial tension, density difference, network height, PSD variance and network coordination number.

It is reasonable to assume that most laboratory measurements of gas flow coefficients (relative permeability, critical gas saturation, residual saturation, etc) relate to CH₄/oil and CH₄/H₂O systems because of their significance in hydrocarbon reservoir engineering. Since reservoir-scale numerical simulation of CO₂ flow processes during sequestration will require properly calibrated CO₂-brine flow coefficients, and given the high cost of laboratory measurements and the apparent similarity between CO₂ and CH₄ (both being gas at standard conditions), there could be a temptation to simply recycle existing CH₄-brine flow coefficients for use in CO₂-brine simulations. At temperature and pressure conditions expected in deep saline aquifers, however, CO₂ would probably exist as a supercritical phase and therefore exhibit very different physical properties from CH₄ (CO₂ is 9 to 10 times denser than CH₄, 30 to 50 times more soluble in brine than CH₄, and 3 to 5 times more viscous than CH₄ above its critical point of 1070.38psia and 31.1°C). Moreover, our limited understanding of the peculiar flow characteristics of CO₂ and CH₄ adds further uncertainty to CO₂-brine simulation results that utilize CH₄/H₂O or CH₄/oil
flow functions. The second part of the chapter therefore examines in detail the differences between the flow behaviours of CO$_2$ and CH$_4$, slowly injected through brine-saturated networks over a range of temperature and pressure conditions. Finally, the implications of the results for reservoir simulation using continuum Darcian models are discussed.

### 4.2 Impact of capillary and gravity forces during gas injection

The key features of the pore network model incorporating the essential physics of gas injection at the pore scale have been presented in the previous chapter. An important pore-level scaling group that quantifies the relative dominance of capillary and gravity forces during injection can be defined through a microscopic Bond number, $B_o$:

$$B_o = \frac{\Delta \rho gh \langle r \rangle}{2\sigma}$$  \hfill (4-1)

where, $\Delta \rho$ is the fluid density difference, $h$ the height of the gas cluster, $\langle r \rangle$ the mean capillary radius, $\sigma$ the interfacial tension, and $g$ the gravitational constant.

Alternative macroscopic definitions are possible, but we shall see that this microscopic form is extremely useful for interpreting the dominant flow regimes. Whilst this Bond number is able to broadly account for some of the governing parameters, others tied to pore architecture – such as the pore size distribution (PSD) variance and coordination number – are not explicitly included. It is shown that these important factors nevertheless exert a strong influence on gravity-driven flow transitions. This is further demonstrated by reproducing two injection experiments, one by Dumoré (1970) and the other by Geistlinger et al (2006).

#### 4.2.1 Model Setup

The porous medium is represented by a 2D lattice of interconnected pore elements and each element is randomly assigned a capillary entry radius from a distribution function with a given mean and variance. Table 4-1 provides a comprehensive list of additional base-case network parameters, including fluid properties and operating conditions.
The system is initially saturated with water of density 1000 kg/m$^3$ and the gaseous phase is injected at a single point close to the bottom of the network at a fixed molar rate. A rate was chosen that yielded a volumetric expansion rate of approximately 0.59E-12 m$^3$/sec (this rate corresponds to an average reservoir scale velocity of approximately 1 m/day, equivalent to injecting at 25 cm$^3$/hour into a 100 cm long core with a diameter of 5 cm and a porosity of 30%). This rate gives a macroscopic capillary number (Ca=qµ/Aσ; q = injection rate, µ = gas viscosity, A = superficial flux area, σ = interfacial tension) of approximately 1.86E-9. The injected phase is considered the non-wetting phase throughout this study.

4.2.2 The Effect of Mean Capillary Radius ($R_{\text{mean}}$)

We begin by examining the effect of mean capillary radius on gas evolution during injection. Twelve different pore size distributions were considered in all, categorised into three groups according to the pore size distribution variance ($a$, $b$, $c$, $d$; $e$, $f$, $g$, $h$; $i$, $j$, $k$, $l$; $\sigma^2 = 6.75$; $\sigma^2 = 200$; $\sigma^2 = 816.75$). The full range of distributions used is shown in Table 4-2.

Figure 4-1 shows snapshots of the developing gaseous phase for all distributions and Figure 4-2 shows the corresponding Bo ($a$, $c$, $e$) plots and saturation profiles ($b$, $d$, $f$). We see that an increase in $R_{\text{mean}}$ leads directly to an increase in the relative impact of gravity forces on the gas cluster morphology. This corresponds to an increase in the microscopic Bond number and a strengthening of flow bias in the vertical direction. Gravitational effects can be observed both within each group of models (e.g. $a$, $b$, $c$, $d$; Figure 4-1) and across model groups ($a$, $e$, $i$; Figure 4-1), as $R_{\text{mean}}$ increased.

For $R_{\text{mean}}$ greater than 75.5 $\mu$m, buoyancy-driven gas mobilization (i.e. migration) is evident (see f, g, h, i, k, and l; Figure 4-1). Migration occurs when $Bo > 1.0$ i.e. when $\Delta \rho gh > 2\sigma/r$ (where $h$ is the height of the associated gas structure) and it is accompanied by water re-imbibition, gas fragmentation and bubble coalescence. The transition from non-migratory to migratory flow is signalled by the change in flow from a progressive elongation of gaseous structures to a more discontinuous flow pattern. On this basis, we can classify the evolution patterns into two broad categories – (a) stable flow and (b)
migratory flow. Each of these can be subdivided into different regimes as discussed below.

### 4.2.2.1 Stable Flow

In the present context, all flows for which \( \text{Bo} < 1.0 \) throughout gas evolution are regarded as stable – stable in the sense that no gas cluster migration occurred. Models \( a, b, c, d, e \) and \( i \) in Figure 4-1 exhibited stable flow behaviour. Two distinct flow regimes could be identified under stable flow.

**Capillary regime:** This is a regime in which gravity forces have little or no effect on the displacement sequence and the injection is completely dominated by capillary pressure conditions. Model \( a \), Figure 4-1, with a maximum Bo of approximately 0.1, Figure 4-5a, lies in this regime.

**Biased Regime:** these are stable regimes in which gravity significantly affects the saturation pattern but no spontaneous migration events occur (Figure 4-1: \( b, c, d, e, \) and \( i \)). The Bo profiles in Figure 4-2a show that, for stable regimes, displacement of the aqueous phase ends once the injected gas breaks through and starts being produced from the top of the system.

The identical saturation patterns of models \( b \) and \( i \) in Figure 4-1 (the pattern in model \( e \) is similar but not identical to the two) is coincidental but also instructive. Gravity bias is governed by a competition between Bond number and the pore network architecture (as represented by the PSD variance and the coordination number). An increase in Bond number increases gravity bias but an increase in PSD variance also supresses it, since the displacement front is now forced to take a more tortuous path. If model \( i \) had the same PSD variance as model \( b \) or a PSD variance just slightly less than its current value, then flow would be more biased in \( i \) than in \( b \) since model \( i \) has a higher average Bond number. Figure 4-3 shows that the flow pattern in model \( i \) can be made more biased by a relatively slight decrease of its current PSD variance, and that there is a short PSD variance range for which model \( i \) will always yield an identical flow pattern as model \( b \). It is apparent that the saturation patterns in \( b \) and \( i \) are identical because the combined effects of Bond number and PSD variance lead to identical *relative* entry threshold distribution fields in
both models. To achieve identical displacement patterns in an invasion percolation process (which stable regimes at low rate essentially are), it is not necessary for absolute entry threshold values at equivalent pore locations in the two networks to be exactly the same. All that is required is that the relative arrangement of high and low entry thresholds in the two networks be identical.

As PSD variance increases (i.e. as the pore size heterogeneity increases) a greater fraction of the total pore volume of the network is held in fewer large pores. Since larger pores are preferentially invaded by gas during displacement, gas saturation builds up quicker as PSD variance increases. This explains why model \(i\) has a higher \(S_{gc}\) than model \(b\) (compare Figure 4-2b[15.5µm] and Figure 4-2f[50.5µm]), even though the endpoint saturation patterns in both are identical. Model \(i\) is more heterogeneous than model \(b\) not just because of its wider PSD variance but also because the ratio between the minimum and the maximum pore size (an alternative measure of heterogeneity) in model \(i\) \((R_{max}/R_{min}=100)\) is 55 times larger than the same ratio for model \(b\) \((R_{max}/R_{min}=1.82)\). The effect of PSD variance on regime transitions is discussed in more detail in section 4.2.5 of this Chapter.

**4.2.2.2 Migratory Flow**

Migratory flow is triggered if \(Bo \geq 1.0\) and involves the spontaneous movement of buoyant gas structures. The models \(f, g, h, j, k,\) and \(l\) in Figure 4-1 display migratory flow behaviour.

A migrating cluster has two active fronts: (a) a leading drainage front, where gas displaces water, and (b) a lagging imbibition front, where water re-imbibes around the base of the migrating gas cluster. The imbibition process is governed by local capillary pressure forces and topological considerations. There are several types of imbibition processes that can occur depending upon the structure of the migrating gas – piston-like advance of the wetting phase, snap off, and intermediate mechanisms (as shown in Figure 3-15, in Chapter 3) – and these mechanisms frequently lead to cluster fragmentation and the formation of a dispersed gas phase consisting of trapped and immobile bubbles.
The classification of regimes under migratory flow, discussed below, is based on the arrangement of these residual bubbles. The Bo and instantaneous gas saturation ($S_g$) profiles may also be used to distinguish one migratory regime from another. One of the main characteristics of migratory flow is that gas saturation continues to change even after gas breakthrough (Figure 4-2: d and f) unlike for stable regimes (Figure 4-2b).

**Braided Regime**

Figure 4-4 presents important stages in the formation of a braided regime. The cluster topology associated with the stable biased regime becomes increasingly elongated as the underlying Bond number (varied here through $R_{\text{mean}}$) increases towards unity. Eventually, the injected gas takes the form of a small number of gravity-biased fingers (and sometimes only a single finger). Now, at some critical $R_{\text{mean}}$, the buoyancy forces begin to exceed the capillary entry thresholds of certain pores around the perimeter of the main gas clusters and the fingers spontaneously migrate. Migration stops as soon as the main structures fragment, as daughter clusters are not sufficiently buoyant to migrate individually. Subsequent injection of gas may either: (i) follow the same path as an old gravity-biased finger and thereby reconnect up the residual bubbles, or (ii) may take a slightly different trajectory depending on the extent to which the previous migration event had perturbed the phase trapping and saturation patterns in the network. Repeated breaking and coalescence events create chains of residual gas clusters that initially follow the paths of the original gaseous fingers. However, new fingers can be initiated and new braided channels develop. The models f and j in Figure 4-1 are typical examples of this behaviour. Bo for braided regimes rises and falls periodically (Figure 4-2: c[125.5µm] and e[100.5µm and 125.5µm]). Each rise is gradual and corresponds to the growth phase, while the fall is sudden and indicates rapid cluster fragmentation during migration.

**Dispersive Regime**

Further increase in $R_{\text{mean}}$ results in a reduction in the critical height threshold required for migration, and migrating clusters are increasingly able to explore and spread to new parts of the network. Figure 4-5 illustrates key stages in the development of dispersive flow. While the fragmenting effects of imbibition may cause intermittent halts to migration, newly injected gas migrating from below may coalesce with stationary clusters and allow
migration to recommence. The sharp spikes of Bo in Figure 4-2c[175.5µm] and Figure 4-2e[250.5µm] highlight these moments of coalescence.

Rapid migration and fragmentation of a gas cluster leaves numerous residual bubbles in its wake and subsequent coalescence of clusters with newly-injected gas can drastically alter the topology of the gaseous phase and hence the distribution of trapped water. Each successive migration thus proceeds on a slightly different path, and ultimately results in a dispersed distribution of trapped bubbles in the network. Hence we can characterise dispersive flow as the development of a residual gas saturation through the repeated trapping and coalescence of cluster fragments along constantly evolving migration pathways. Models g, h, k, and i in Figure 4-1 exhibit dispersive behaviour.

It is clear that there are two necessary conditions for dispersion of an injected gas phase to occur:

1. A large microscopic Bond number such that $\Delta \rho gh > 2\sigma/\langle r \rangle$ (where h is the height of the associated gas structures)
2. Substantial cluster fragmentation during migration

Of course, we do not know the height of individual gas structures during any given experiment a priori and pore-scale simulation provides one of the few available tools to predict this important behaviour. Figure 4-6 summarises typical saturation patterns corresponding to the flow regimes discussed so far and Figure 4-7 shows the associated Bo signatures.

Having broadly outlined the significance of the changes in the relative dominance of capillary and gravity forces to gas injection flow regimes solely by varying the mean capillary radius, we now go on to examine the influence of other parameters that control capillary and gravity forces in more detail.
Table 4-1: Summary of Network parameters, fluid properties and operating conditions

<table>
<thead>
<tr>
<th>Fluids:</th>
<th>Water and gas</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial Pressure, [MPa]; [psi]</td>
<td>5.17; 750</td>
</tr>
<tr>
<td>Temperature, [$^\circ$C]</td>
<td>35</td>
</tr>
<tr>
<td>Gravity:</td>
<td>Always active</td>
</tr>
<tr>
<td>Injection Rate, [m$^3$/sec]</td>
<td>0.59e-12</td>
</tr>
<tr>
<td>Gas Dissolution:</td>
<td>Off</td>
</tr>
<tr>
<td>Z</td>
<td>6</td>
</tr>
<tr>
<td>IFT, [mNm$^{-1}$]</td>
<td>15.0</td>
</tr>
<tr>
<td>NX, NY, NZ</td>
<td>131, 75, 1</td>
</tr>
<tr>
<td>Pore length, [µm]</td>
<td>300.0</td>
</tr>
<tr>
<td>Total model height [cm]</td>
<td>3.93</td>
</tr>
<tr>
<td>Buffer</td>
<td>3 bond rows</td>
</tr>
<tr>
<td>Water density [kg/m$^3$]</td>
<td>1000</td>
</tr>
<tr>
<td>Gas density [kg/m$^3$]</td>
<td>38</td>
</tr>
</tbody>
</table>

Table 4-2: Pore Radii distributions for three groups of models (abcd; efgh; and ijkl) arranged according to PSD variance

<table>
<thead>
<tr>
<th>Sigma$^2$</th>
<th>6.75</th>
</tr>
</thead>
<tbody>
<tr>
<td>R$_{\text{mean}},$ µm</td>
<td>5.5 15.5 25.5 35.5</td>
</tr>
<tr>
<td>R$<em>{\text{min}}$, R$</em>{\text{max}},$ µm</td>
<td>1, 10 11, 20 21, 30 31, 40</td>
</tr>
<tr>
<td>Sigma$^2$</td>
<td>200</td>
</tr>
<tr>
<td>R$_{\text{mean}},$ µm</td>
<td>25.5 75.5 125.5 175.5</td>
</tr>
<tr>
<td>R$<em>{\text{min}}$, R$</em>{\text{max}},$ µm</td>
<td>1, 50 51, 100 101, 150 151, 200</td>
</tr>
<tr>
<td>Sigma$^2$</td>
<td>816.75</td>
</tr>
<tr>
<td>R$_{\text{mean}},$ µm</td>
<td>50 100 150 250</td>
</tr>
<tr>
<td>R$<em>{\text{min}}$, R$</em>{\text{max}},$ µm</td>
<td>1, 100 51, 150 101, 200 201, 300</td>
</tr>
</tbody>
</table>
### Figure 4-1: The impact of mean capillary radius ($R_{\text{mean}}$) on regime transitions. Models arranged according to the layout of Table 4-2.

<table>
<thead>
<tr>
<th>$\sigma^2$</th>
<th>$R_{\text{min}}, R_{\text{max}}$ [\mu m]</th>
<th>a</th>
<th>b</th>
<th>c</th>
<th>d</th>
</tr>
</thead>
<tbody>
<tr>
<td>6.75</td>
<td>1, 10</td>
<td>11, 20</td>
<td>21, 30</td>
<td>31, 40</td>
<td></td>
</tr>
<tr>
<td>200.0</td>
<td>1, 50</td>
<td>51, 100</td>
<td>101, 150</td>
<td>151, 200</td>
<td></td>
</tr>
<tr>
<td>816.75</td>
<td>1, 100</td>
<td>51, 150</td>
<td>101, 200</td>
<td>201, 300</td>
<td></td>
</tr>
</tbody>
</table>
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Figure 4-2: Sensitivity of instantaneous maximum local Bo measured over all gas clusters (Bo_max) and instantaneous gas saturation, S_g, profiles to R_mean for the models in Figure 4-1. (a, b) σ^2 = 6.75, (c, d) σ^2 = 200.0, (e, f) σ^2 = 816.75. To determine Bo_max, the local Bo at each water-filled pore at the perimeter of a gas cluster is calculated according to Equation (4-1) – where h is in this case the vertical height of the pore from the base of the gas cluster – and sorted. The maximum value of the local Bos is taken as Bo_max.

<table>
<thead>
<tr>
<th>Sigma^2</th>
<th>816.75</th>
<th>784.083</th>
<th>720.75</th>
<th>660.083</th>
<th>602.083</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1, 100</td>
<td>2, 99</td>
<td>4, 97</td>
<td>6, 95</td>
<td>8, 93</td>
</tr>
</tbody>
</table>

Figure 4-3: Impact of PSD variance on displacement pattern of model of i in Figure 4-1. 1, 100; 2, 99;... denote corresponding R_min and R_max values in µm.
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Figure 4-4: Stages in the formation of a braided regime in model f in 4-1. Note how the residual saturation distribution (phase trapping patterns) changes with each growth-migration cycle (a1 – a4. The phase trapping pattern perturbations eventually lead to the creation of new evolution routes.

Figure 4-5: Key moments in the development of a dispersive regime for model k in Figure 4-1; Note: b and c show trajectory deflections induced by phase trapping pattern changes; d, f, and h, show coalescence events; e, g and i, show the aftermath of the coalescence events of d, f, and h, respectively; j shows a trajectory change, and k its aftermath; the end point saturation distribution is shown in l.
4.2.3 Effect of Interfacial Tension and Density Difference

The next part of the study focuses on the impact of gas-water interfacial tension (IFT) upon gas injection. Since Bond number is inversely proportional to IFT, a decrease in IFT implies an increase in Bond number and a reduction in the influence of capillary forces. Here, network $R_{\text{mean}}$ and PSD variance were maintained at 5.5 microns and 6.75, respectively. As the IFT was decreased from 90 mN/m to 0.069 mN/m we move through the full range of gas flow regimes: a capillary dominated regime at high IFT and a dispersive migratory at low IFT, and in between the two extremes there is a gravity biased regime (Figure 4-8). Figure 4-9a shows the trend in $S_{gc}$ as IFT was varied – the high $S_{gc}$ at the opposite extremes of IFT values straddle a low $S_{gc}$ at an intermediate IFT value.
As expected, an increase in $\Delta \rho$ increased the gravity bias in the same way as an increase in $R_{\text{mean}}$ or a decrease in IFT. Critical saturation decreased with increase in $\Delta \rho$. The monotonic trend of the $S_{gc}$ vs $\Delta \rho$ curve in Figure 4-9b was due to the short Bo range over which the $\Delta \rho$ sensitivity spanned. No transition to the migratory regime occurred over the $\Delta \rho$ change, unlike for the IFT sensitivity in Figure 4-9a.

4.2.4 Impact of Length Scale
For completeness, the impact of network height on regime transition was examined. Four different network heights were considered (2cm, 4cm, 8cm, and 20cm) and a pore size distribution with $R_{\text{mean}}$ of 25µm ($\sigma^2 = 6.75$) was used. The pore length and network aspect ratio were also kept constant for all simulations.

Figure 4-10 shows gas flow transition from a capillary-dominated displacement, through gravity-biased fingering, to a braided migratory regime and finally a more dispersive regime. A non-monotonic trend in the plot of $S_{gc}$ against model height is evident (Figure 4-11) – viz: a continuous decrease in $S_{gc}$ that accompanies a regime transition from stable capillary dominated flow to a stable gravity biased regime is followed by a small increase
in $S_{gc}$ as the flow regime becomes migratory and dispersive. The implications of this trend to the choice of core sizes and core orientation during laboratory determination of two-phase flow functions will be discussed later.

**Model Height, [cm]; ($R_{\text{mean}} = 25 \ \mu m$)**

<table>
<thead>
<tr>
<th>2</th>
<th>4</th>
<th>8</th>
<th>20</th>
</tr>
</thead>
</table>

Figure 4-10: The impact of network height on regime transition

Figure 4-11: The impact of network height on $S_{gc}$ for a constant $R_{\text{mean}}$ of 25.5microns.
4.2.5 Effect of PSD Variance ($\sigma^2$)  
We now move on to investigate the impact of parameters related to the network microstructure, starting with the pore size distribution variance. The goal is to vary $\sigma^2$ whilst $R_{\text{mean}}$ fixed, the opposite of the situation in Figure 4-1, where $R_{\text{mean}}$ was varied at $\sigma^2$ values. Increasing the PSD variance increases capillary pressure heterogeneity and Table 4-3 gives the details of the models selected for this sensitivity. The models were listed under two groups, “a, b, c, d” and “e, f, g, h”, according to $R_{\text{mean}}$ (25.5µm and 250.5µm). These $R_{\text{mean}}$ values were chosen to demonstrate the effect of PSD variance on both stable and migratory regimes. The difference between models in each group was their PSD variance. Although we choose results from networks with relatively large pore sizes, similar results can be envisaged for smaller pores coupled with lower interfacial tensions.

**Stable Regimes**

As can be observed in Figure 4-12(a, b, c, & d), gravity fingers become more biased as $\sigma^2$ decreases. At high $\sigma^2$, the local pore entry thresholds are widely distributed and the injected gas flowed through a slightly perturbed capillary-dominated path. Conversely, when $\sigma^2$ is small, the local hydrostatic pressure becomes the main perturbing force. Water pores near the top of the growing cluster are preferentially drained, resulting in more directed growth patterns.

**Migratory Regimes**

**Dispersive**: The models in Figure 4-12(e, f, g, & h) are all dominated by dispersive regimes. We find that the degree of network heterogeneity determines the degree of dispersion during injection. Gas injected into a more heterogeneous medium (high $\sigma^2$) fragments into numerous dispersed clusters, whilst more compact, slug-like structures dominate in a homogeneous (low $\sigma^2$) medium (Figure 4-13-i). During migration, these compact clusters are re-imbibed predominantly via piston-like displacements at their lower edge, yielding very few residual bubbles in their wake. $S_{gc}$ increases considerably as the pore size distribution widens (Figure 4-13-ii).
Table 4-3: Pore Radii distributions for the two (abcd and efgh) groups of models used for PSD variance sensitivity, arranged according to $R_{\text{mean}}$.

<table>
<thead>
<tr>
<th>$R_{\text{mean}}, \mu$m</th>
<th>25.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Sigma^2$</td>
<td>0.75</td>
</tr>
<tr>
<td>$R_{\text{min}}, R_{\text{max}}, \mu$m</td>
<td>a</td>
</tr>
<tr>
<td>-------------------------</td>
<td>------</td>
</tr>
<tr>
<td>$R_{\text{mean}}, \mu$m</td>
<td>250.5</td>
</tr>
<tr>
<td>$\Sigma^2$</td>
<td>200</td>
</tr>
<tr>
<td>$R_{\text{min}}, R_{\text{max}}, \mu$m</td>
<td>e</td>
</tr>
</tbody>
</table>

Figure 4-12: The impact of pore size distribution variance ($\sigma^2$) on regimes, arranged according to the layout of Table 4-3.

Figure 4-13: (i) Comparison of typical migrating cluster morphologies in (a) Heterogeneous (Figure 4-12h) and (b) Homogeneous (Figure 4-12e) networks; (ii) The impact of PSD variance upon $S_{gc}$ for a dispersive migratory regime ($R_{\text{mean}}=250\mu$m).
4.2.6 Effect of Coordination Number
A relatively homogeneous (low $\sigma^2$) model was selected for this sensitivity study and the discussion will focus on migratory regimes for brevity.

Figure 4-14 shows that a decrease in coordination number (Z) has a similar effect on dispersion as an increase in PSD variance – reduced Z produces dendritic clusters highly prone to fragmentation and dispersion. Reduced connectivity thus magnifies the degree of dispersion during gas injection. The obstructive role played by the smaller pores in a system with a large variance in pore-size is analogous to the reduced availability of open pore space in low Z systems. *It should be noted that because of the restrictive nature of 2D networks, the observed effect of a decrease in Z on flow might have been exaggerated. Simulations using 3D network models would provide a better quantitative measure of the effect of Z on flow behaviour.*

![Figure 4-14: The impact of Coordination number on Migratory regimes.](image)

<table>
<thead>
<tr>
<th>2D, Z</th>
<th>4.0</th>
<th>3.33</th>
<th>2.67</th>
<th>2.33</th>
</tr>
</thead>
<tbody>
<tr>
<td>$R_{\text{mean}}$ = 250.5 µm</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\sigma^2$ = 200</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

4.2.7 Impact of Injection Configuration
So far, we have only considered gas injected at a single point in the system. Now we consider injecting across the entire bottom face of the network (at the same molar injection rate). Four models corresponding to each of the four main flow regimes found under single point injection were considered. Figure 4-15 shows that the resulting flow regimes were unaffected by injection configuration.
4.2.8 Comparisons of the injection experiments of Dumoré (1970) and Geistlinger et al, (2006)

We next compare the model formulation against available experimental observations and present examples of the high sensitivity of gravity-driven regimes to the underlying microstructure of the porous medium – modelled here through the variance of the pore size distribution (PSD) and pore connectivity. We begin by considering the dispersion and the non-dispersion injection experiments of Dumoré (1970) reproduced in Figure 4-16.

Here, gas was injected at a low rate into two oil-filled bead packs of different permeabilities such that viscous forces were negligible. Although permeability values were not provided by Dumoré, they were estimated to be ~150 Darcies and ~90 Darcies for the coarse and fine packs respectively. These values were reached by using the reported pack capillary pressures, fluid interfacial tension, and porosities, according to: $K = \phi / (Pc/\sigma)^2$, where $\phi$, $Pc$, $\sigma$ are the porosity, capillary pressure and interfacial tension, respectively. The permeability values were then converted to mean pore radii using: $R_{mean} = \sqrt{(32K/\phi)/2}$ (i.e. via a simple capillary bundle approximation).

Two dimensional networks of size 131 x 75 nodes were used and a pore length of 230µm was inferred from the available data. However, the in silico networks represent a total height of 3cm, whilst Dumoré’s bead packs spanned 54cm and so the gravitational constant in our model was scaled in order to match experimental Bond numbers. The injection fluxes corresponding to the network model and the experimental bead packs

<table>
<thead>
<tr>
<th>$\bar{R} [\mu m]$</th>
<th>5.5</th>
<th>35.5</th>
<th>75.5</th>
<th>250.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>Single point injection</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Full face injection</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Capillary</td>
<td>Biased</td>
<td>Braided</td>
<td>Dispersive</td>
<td></td>
</tr>
<tr>
<td>Stable</td>
<td>Migratory</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 4-15: The impact of injection configuration of regimes.
were also matched. For an injection rate of 5cc/day (or 5.79E-11 m³/sec) used in the experiment, the scaled injection rate used in the model was calculated to be 7.63E-03cc/day (or 8.84E-14 m³/sec).

\[ a - \text{coarse} \quad b - \text{fine} \]

Figure 4-16: The (a) dispersion and (b) non-dispersion injection experiments of Dumoré (1970).

Figures 4-17 and 4-18 show the results of the simulations and clearly demonstrate the strong link between gas dispersion and pore microstructure. As the microstructure becomes more heterogeneous through either an increase in PSD variance, or a decrease in pore connectivity, flow becomes more dispersed. They also show that the differences in absolute permeability between the two packs may not be as fundamental a control as the differences in their underlying architecture for determining whether dispersion occurs. The dispersion and non-dispersion results of Dumoré can be reproduced by using either the high or low permeability networks through variations in PSD variance and/or the coordination number. Hence, we can conclude that absolute permeability is not a sufficiently robust parameter for inferring the onset of dispersive flow during gas injection into a porous medium.

<table>
<thead>
<tr>
<th>A</th>
<th>Heterogeneous -&gt; Homogeneous</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fine Pack</td>
<td><img src="image1" alt="Image" /></td>
</tr>
<tr>
<td>( R_{\text{mean}} = 43 , \mu m )</td>
<td><img src="image2" alt="Image" /></td>
</tr>
<tr>
<td>( R_{\text{min}}, R_{\text{max}} )</td>
<td>25, 60</td>
</tr>
<tr>
<td>B</td>
<td>Coarse Pack</td>
</tr>
<tr>
<td>( R_{\text{mean}} = 56 , \mu m )</td>
<td><img src="image3" alt="Image" /></td>
</tr>
<tr>
<td>( R_{\text{min}}, R_{\text{max}} )</td>
<td>31, 81.6</td>
</tr>
</tbody>
</table>

Figure 4-17: Simulation of the impact of PSD variance on regimes in the (A) Fine and (B) Coarse packs of Dumoré (1970). Coordination number = 4.
Chapter 4 - Gravity Destabilised Gas Flow Regimes

### Table 4-1

<table>
<thead>
<tr>
<th>2DZ</th>
<th>2.67</th>
<th>3.33</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fine Pack</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$R_{\text{min}}, R_{\text{max}}$</td>
<td>40, 45</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Coarse Pack</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$R_{\text{min}}, R_{\text{max}}$</td>
<td>51, 61.6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Figure 4-18: Simulation of the impact of coordination number on regimes in the (A) Fine and (B) Coarse packs of Dumoré (1970).

An analogous scaling argument was used to qualitatively reproduce results of the single-point gravity-destabilised injection experiments of Geistlinger et al (2006). The experiment used three separate glass bead packs characterised by different bead size distributions (0.25-0.50mm denoted as 0.5mm-GBS; 0.75-1.0mm denoted as 1mm-GBS; and 2.0-2.2mm denoted as 2mm-GBS). Gas (air) was injected near the bottom of each pack at a rate of approximately 10mL/min.

Figure 4-19 shows the PSD in each pack as given in Geistlinger et al (2006). 2D network models with $N_x*N_y*N_z$ node dimensions of 131x117x1 were used for these simulations. An average pore was assumed to be 228µm long and this implies a total network height of approximately 3cm, a width of 2.7cm and thickness equal to 0.0228cm – these dimensions maintain the same height:width aspect ratio as the experimental bead packs which measured 45cmx40cmx1.2cm.

The gravitational constant in the model was modified by multiplication with a scaling factor defined by $\text{Height}^{\text{experiment}}/\text{Height}^{\text{model}} = 15$. The equivalence of an experimental flow rate of 10mL/min ($1.67E-07 \text{ m}^3/\text{sec}$) was found to be $2.11E-09 \text{ m}^3/\text{sec}$ or 0.13mL/min in the network via comparison of the effective flux areas of the pack and the model. An air-water interfacial tension of 72Nm$^{-1}$, at a temperature and pressure of 25°C and 0.103MPa respectively, was applied in both the network model and the experimental bead packs. The macroscopic capillary number ($Ca = Q\mu/\sigma A$) for the experiment was
calculated as approximately 8.90E-09 using an air viscosity of 2e-5PaS. This justifies our assumption of ignoring any effects of viscous forces.

Figure 4-19 compares the pore occupancy graphics from simulation results – performed using a coordination number of 4 – and the experimental results. Our simulations correctly predict the transition from braided migration to dispersive flow as mean bead size increases.

Figure 4-19: Comparison of lab (Geistlinger et al, 2006) and simulation results of regime transitions with change in mean pore radius.

<table>
<thead>
<tr>
<th>Model</th>
<th>0.5mm</th>
<th>1mm</th>
<th>2mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>R_{mean}, μm</td>
<td>85</td>
<td>210</td>
<td>480</td>
</tr>
<tr>
<td>R_{min}, R_{max}, μm</td>
<td>57, 113</td>
<td>140, 280</td>
<td>320, 640</td>
</tr>
</tbody>
</table>

4.2.9 Implications of Gravity-Driven Regime Transitions

We briefly highlight some implications of the influence of regime transitions upon critical gas saturation in response to a wide range of fluid and network parameters. Here, critical gas saturation is simply the fraction of the total network pore volume occupied by gas at the point of first breakthrough and would thus correspond to the first value of saturation with a non-zero relative permeability on a relative permeability versus saturation curve. The definition applies equally to migratory regimes, where we can monitor discontinuous gas production at the outlet.

4.2.9.1 R_{mean}, IFT, Δρ, and Length Scale

The separate investigations of the impact of R_{mean}, IFT, Δρ, and length-scale on regime transitions are in effect an investigation of the impact of Bond number (Bo) upon gravity-driven flow patterns. We find that S_{gc} initially decreases from its maximum value (which
occurs under capillary-dominated conditions) as $Bo$ increases towards 1 (as we move from a capillary regime ($0 < Bo \leq 0.1$), to a biased regime ($0.1 < Bo < 1$)). For $Bo \geq 1$, $S_{gc}$ begins to increase again as the regime becomes migratory and we find that the extent of this increase depends on the specific porous medium architecture – the more heterogeneous and disconnected it is, the greater the increase in $S_{gc}$.

4.2.9.1.1 Parameterization of continuum models
It is generally considered best practice to minimise the influence of gravity forces during laboratory determination of multiphase flow parameters for use in continuum models, e.g. relative permeability and $S_{gc}$, and this is usually achieved by conducting displacement experiments using horizontally oriented cores. Any gravitational effects on flow during simulation are then accounted for through the incorporation of gravity forces in the underpinning Darcian flow equations. Given that a typical reservoir simulation gridlock is several orders of magnitude larger than an average experimental core, different flow regimes might be active at the two scales, with the core-scale regime more likely to be capillary dominated and stable, and the gridblock-scale regime migratory. If the change in regime due to a change in the scale of reference is only a matter of degree, then the modification to multiphase flow functions during upscaling might be relatively straightforward. It would involve, as a first approximation, a shift of the relative permeability endpoints and a preferential adjustment of interblock mobility in the direction of the dominant gravitational gradient, in effect making $S_{gc}$ a directional parameter. At present, there is no reported upscaling procedure for accounting for such a change in regime during reservoir-scale simulation of gravity dominated flow processes such as CO$_2$ storage.

If a change of scale resulted in a transition from a stable regime to a migratory regime, then a more sophisticated procedure for upscaling relative permeability that goes beyond simply shifting the endpoints may be required. This is because a stable to migratory regime transition involves a significant change in the saturation distribution – CO$_2$ is now more disconnected and dispersed and therefore presents larger CO$_2$-water surface areas for dissolution. Furthermore, the dynamic evolution of the residual saturation in the migratory regime must also be accounted for and recent studies that use Darcian simulators to evaluate residual trapping capacity during CO$_2$ storage, by the use of
hysteretic relative permeabilities (Ehsan et al, 2008; Juanes et al, 2008) provide a useful
starting point.

Gravity-driven regimes are not only sensitive to length scale, however. The strong
temperature and pressure dependence of IFT and $\Delta \rho$ emphasises the need for laboratory
experiments to be carried out as near to the conditions of the intended formation as
possible. Furthermore, since it is common to expect fairly wide variations of rock
properties even within small regions of a typical reservoir, there arises the familiar
question of what size of sample is truly representative. Our results suggest that an
increase in mean pore radius (a correlate of permeability) by a factor of less than 2 – e.g.
from 15.5microns to 25.5microns, could decrease $S_{gc}$ by more than 50%. Hence, in order
to sufficiently reduce the uncertainty in parameterization, rigorous and robust sampling
strategies will be required. Network modelling could ease these practical constraints by
allowing several parametric sensitivities to be performed in a timely manner to yield
information about the kind of regimes to expect.

4.2.9.2 PSD Variance and Coordination Number (Z)

The in silico results have shown that $S_{gc}$ increased with $\sigma^2$ for both stable and migratory
regimes. The effect of Z on gas evolution presents a scenario where the impact depends
on whether the regime is stable or migratory. A decrease in Z might lead to severe
meandering in stable regimes but the changes to $S_{gc}$ are statistically negligible. For
migratory regimes on the other hand, we see a sharp jump in $S_{gc}$ followed by a decline as
Z decreases.

4.2.9.2.1 Parameterization of continuum models

The impact of core-scale rock heterogeneities due to stratification on reservoir-scale flow
behaviour is well known and methodologies have been developed to account for these
during upscaling of macroscopic flow parameters, (Rustad et al, 2008; Lohne et al, 2004;
Crotti and Cobenas, 2001; and Saad et al, 1995). Our results show that heterogeneity in
pore size ($\sigma^2$) and pore connectivity could have equally strong effects.

The impact of $\sigma^2$ on gravity-driven regimes reveals the limitations of the conventional
definition of Bo as a reliable indicator of the degree of gravity bias. This also means that
the conventional Bo cannot be satisfactorily used as a scaling parameter without incorporating the effects of pore-scale heterogeneity.

4.3 Comparison of CH₄-H₂O and CO₂-H₂O Binary Fluid Systems

The classification of gas evolution patterns into different migration regimes provides a useful insight into the way in which gas structures evolve during injection. Moreover, the flow regime scheme described above could also be used as a practical screening tool for the reuse of existing multiphase flow functions (e.g. relative permeability and Pc functions) for reservoir simulation studies where new experiments cannot be easily performed – due to a shortage of physical samples or because experimental equipment is unable to handle the specific temperature and pressure conditions of interest. An example might be the use of relative permeability data derived from a methane-oil or a methane-brine injection experiment for use in a CO₂-brine injection simulation. To illustrate, we now demonstrate an application of the model by comparing flow regimes obtained by using CH₄-H₂O and CO₂-H₂O binary fluid systems, under various combinations of capillary and gravity forces and at different conditions of temperature and pressure.

The system temperature and pressure values for the first set of simulations, 35°C and 10.34MPa, respectively, were selected to be slightly above the supercritical point of CO₂ (7.38MPa, 31.1°C) and the corresponding fluid and rock/fluid properties at these conditions are listed in Table 4-4. *Note that no dissolution of gas into the brine was considered at this stage.* The gravitational constant was appropriately scaled to allow the full spectrum of gravity-driven regimes to be examined with a network height of 2cm (73 x 37 x 1 nodes). Details of the network pore size distribution can be found in Table 4-2.

Figure 4-20 compares pore occupancy graphics obtained by injecting CH₄ and CO₂ at equal volumetric rates of 0.295E-12m³/sec into initially 100% water-saturated networks. The resulting CH₄ and CO₂ flow regimes in equivalent networks were found to be broadly the same (the only exception were the results obtained from networks with R_mean=75µm, where changing the injected gas from CH₄ to CO₂ led to a switch from a migratory regime to a stable regime).
In changing the injected phase from CH$_4$ to supercritical CO$_2$, multiple model parameters are altered simultaneously – IFT, $\Delta \rho$, viscosity, solubility, and diffusion coefficient. Because gas dissolution was disabled and viscous forces were negligible, any differences between CH$_4$ and CO$_2$ saturation profiles in equivalent networks can only be accounted for by the discrepancy between CH$_4$/H$_2$O and CO$_2$/H$_2$O IFTs and density differences. From Table 4-4, IFT$_{\text{CH}_4/\text{H}_2\text{O}} \approx 2\text{IFT}_{\text{CO}_2/\text{H}_2\text{O}}$, while $\Delta \rho_{\text{CH}_4/\text{H}_2\text{O}} \approx \Delta \rho_{\text{CO}_2/\text{H}_2\text{O}}/3$, and so CH$_4$ flow had slightly larger Bond numbers than CO$_2$ flow and was therefore more gravity biased than CO$_2$ (see Figure 4-21). Because the difference in Bo between equivalent CH$_4$-H$_2$O and CO$_2$-H$_2$O models was relatively small, Figure 4-22 shows that the differences between CO$_2$ and CH$_4$ saturation profiles in equivalent networks were also small so long as the system was away from the stable/migratory crossover point (see Figures 4-21-iii and 4-22-iii). 

**Note that, when the systems were close to Bo=1, even small perturbations in the fluid properties could substantially affect the build-up of the injected gas.**

### 4.3.1 Impact of Gas Dissolution

Up until this point, dissolution of the injected gas in water has not been considered. We now go on to consider the impact of diffusive mass transfer during injection on the evolving saturation distributions.

Dissolution is modelled here through Fickian diffusion, whereby dissolved gas concentrations and the molar gas content of individual gas structures are updated dynamically at each timestep (see Bondino et al, 2005 and Ezeuko et al, 2009 for details). CO$_2$ solubility was calculated using the model of Duan et al (2006), whilst CH$_4$ solubility values were taken from the NIST online database (solubility was expressed in terms of kilograms of solute (CO$_2$) per m$^3$ of solvent (H$_2$O)). The amount of dissolved CO$_2$ in brine is always in flux due to continuous diffusive mass transfer from CO$_2$ to brine, at a rate determined according to

$$\dot{m}_{ij} = D(C_j - C_i) \times \text{min}(A_i, A_j)/L \quad \text{(4-2)}$$

where, $\dot{m}_{ij}$ = mass flux from pore $i$ to pore $j$ [kg/sec], $D$ = diffusion coefficient [m$^2$/sec], $L$ = a diffusion length [m], taken as the distance between two pore
centres, \( C \) is the concentration [kg/m\(^3\)] and \( \min(A_i, A_j) \) is the minimum of the cross-sectional areas of pore \( i \) and pore \( j \).

As the free gas phase loses mass to the aqueous phase during dissolution, an algorithm is required to adjust (i.e. shrink) the current volume of free gas to satisfy the new thermodynamic conditions. This is achieved by locally re-imbibing the requisite volume of water around the shrinking gas structures – local capillary pressures (\( P_c \)), net hydrostatic pressures and network topology are all taken into account (according to Equation 3-15, described earlier in Chapter 3).

The dissolved \( \text{CO}_2 \) concentration of subsurface brines \textit{in situ} may range from a negligible value to complete supersaturation depending on a number of factors which include their hydrodynamic histories – prior or existing contact with a natural \( \text{CO}_2 \) vent or with \( \text{CO}_2 \) enriched surface waters, and the mineralogy of the host formation. Here, the initial dissolved \( \text{CO}_2 \) concentration in brine was set to 10% of the equilibrium concentration at the modelled temperature and pressure conditions i.e. brine was initially 90% undersaturated.

Figure 4-23 and Figure 4-24 show the effect of gas dissolution in brine during the injection of \( \text{CH}_4 \) and \( \text{CO}_2 \), respectively. These show that gas dissolution had a smaller impact on \( \text{CH}_4 \) saturation profiles than \( \text{CO}_2 \) profiles because of the lower solubility of \( \text{CH}_4 \) in \( \text{H}_2\text{O} \) (see Table 4-4). Dissolution slows down the build-up of gas and reduces the saturation at which gas breakthrough occurs. The degree of this delay increases with increase in \( R_{\text{mean}} \) and diffusion surface area. The water re-imbibition process that contracts the free gas structures during dissolution may also have the secondary effect of altering the gas structure topology, such that subsequent drainage displacement paths may deviate from the previously established path – possibly resulting in an increase or a decrease in breakthrough saturation. \textit{Nonetheless, even after the effect of gas dissolution is taken into account, \( \text{CH}_4 \) and \( \text{CO}_2 \) saturation profiles in equivalent networks remain very similar when the \( \text{CO}_2 \) temperature and pressure are above the supercritical point} (Figure 4-25). Changes in evolution pathway due to \( \text{CO}_2 \) dissolution is shown in Figure 4-26.
4.3.2 Comparison of CH₄-H₂O and CO₂-H₂O Systems Away from the Critical Point of CO₂

The three main parameters that differentiate a CH₄-H₂O system from a CO₂-H₂O system are IFT, Δρ, and solubility and each is a function of temperature and pressure. The aim here is to discover to what extent changes in these parameters due to varying temperature and pressure conditions would impact on breakthrough (i.e. critical) saturations of CO₂ and CH₄.

Four additional conditions of temperature and pressure combinations were considered in addition to the first condition at 35°C and 10.34MPa. The conditions and their acronyms are given as follows:

1. T=35°C (critical) and P=10.34MPa (critical). This condition will be denoted as \((TP)_{crit}\)
2. T=25°C (subcritical) and P=0.69MPa (subcritical). Denoted as \((TP)_{sub}\)
3. T=25°C (subcritical) and P=24.13MPa (supercritical). Denoted as \(T_{sub}P_{super}\)
4. T=100°C (supercritical) and P=0.69MPa (subcritical). Denoted as \(T_{super}P_{sub}\)
5. T=100°C (supercritical) and P=24.13MPa (supercritical). Denoted as \((TP)_{super}\)

Figure 4-27 compares fluid properties at the five temperature and pressure conditions considered. Although all five fluid properties changed with varying conditions, it was the changes in IFT, Δρ, and solubility which will be shown to exert the most significant impact on flow.

4.3.2.1 Estimation of Dissolution Rate

The simulations at \((TP)_{crit}\) have demonstrated that gas (CO₂ or CH₄) dissolution in brine had minimal effect on the structure of the free gas phase. We will now examine the dissolution rate and dissolution capacity of brine to CO₂ and CH₄ at the five temperature and pressure conditions selected in more detail.

The network parameters remain essentially the same as in the last section but with a few modifications – gravity force was deactivated, initial aqueous CO₂ concentration was set at zero, and after 0.15PVI (of CH₄ and CO₂), injection was stopped and the gas allowed sufficient time to dissolve. Dissolution was deactivated prior to shut-in to further simplify analysis.
Figure 4-28 shows comparisons of the saturation profiles of CO$_2$ and CH$_4$ as functions of shut-in time for a network $R_{\text{mean}}$ of 75$\mu$m, at five temperature and pressure conditions. It is clear that dissolution rate was strongly dependent on temperature and pressure and that CO$_2$ dissolved more readily than CH$_4$, as a consequence of its higher solubility. Furthermore, the dissolvability of brine to gas – defined here as the volume of brine required to dissolve a unit volume of gas at a given temperature and pressure – was inversely proportional to system pressure (CO$_2$ and CH$_4$ dissolved quicker at 0.69MPa $[(TP)_{\text{sub}} & T_{\text{super}P_{\text{sub}}}]$ than at 10.34MPa and above $[(TP)_{\text{crit}}, (TP)_{\text{super}}, T_{\text{sub}P_{\text{super}}}]$). This means that as pressure increases the fraction of the free gas that can dissolve in brine decreases even as the gas solubility increases. We must remember that just as solubility increases with pressure so does the gas density. Figure 4-29 and Figure 4-30 show that density increases with pressure at a faster rate than solubility, especially once the supercritical boundary has been crossed. Thus, a greater mass of free gas would have to go into solution at a higher pressure than at a lower pressure in order to reduce the free gas volume by the same amount at both conditions.

Since the evaluation of storage capacity and flow behaviour is ultimately based on proper tracking of fluid volumes rather than mass, we propose a dissolvability index to help better indicate the ability of brine to sequester CO$_2$ or CH$_4$ in the aqueous phase under changing temperature and pressure conditions. Equation 4-3 is a simple expression for calculating such an index – as the ratio of solubility of phase $i$ (e.g. CO$_2$) in phase $j$ (e.g. brine) to the density of phase $i$, all evaluated at the same temperature and pressure.

$$D^{ij}(T, P) = \frac{S^{ij}(T, P)}{\rho_i(T, P)}$$

(4-3)

where $D^{ij}(T, P)$ is the dissolvability of phase $i$ (free CO$_2$ or CH$_4$) in phase $j$ (aqueous brine); $S^{ij}(T, P)$ the solubility of phase $i$ in phase $j$ [kg/m$^3$]; and $\rho_i(T, P)$ the density of phase $i$ [kg/m$^3$], all functions of temperature and pressure. Figure 4-31 shows a temperature-pressure phase diagram of the dissolvability index.
To return to our simulation results, Figure 4-32 compares the correlation of total dissolved CO₂ (as a fraction of network PV) to the absolute solubility (expressed as \( K_{\text{CO}_2} / K_{\text{H}_2\text{O}} \)) and the dissolvability index \( (m_{\text{CO}_2}^3 / m_{\text{H}_2\text{O}}^3) \), at the five temperature and pressure conditions. It confirms the trends observed in Figure 4-31, and demonstrates that, compared to the absolute solubility, the dissolvability index might be a better predictor of the volumetric fraction of injected gas than can be dissolved under any given condition and this implies that supercritical CO₂ could be less dissolvable in brine than current understanding suggests.

Starting with a total CO₂ PVI of 0.15, only 0.057PVI was dissolved at the condition \((TP)_{\text{crit}}\) before brine became completely saturated, whereas for the \((TP)_{\text{sub}}\) condition all free CO₂ went into solution before brine became saturated (also see Figure 4-33). The implication of this for CO₂ sequestration is that at the temperature and pressure conditions which prevail in deep saline aquifers the fraction of CO₂ that can ultimately dissolve is, in volumetric terms, limited to the equivalent of about 0.06 of aquifer PV (pore volume). Any extra CO₂ volume above 0.06PV would likely remain as a free phase, potentially mobile or trapped by capillary forces. Moreover although the back production of aquifer brine could free up more pore volume for CO₂ injection and increase storage capacity, it may also diminish storage security – as less brine is available into which CO₂ can be dissolved and ultimately mineralised. Hence, it is crucial to ensure that injection strategies are optimized to maximize residual trapping of free CO₂.

Figure 4-34 shows the impact of dissolution on free CO₂ structures with time at a supercritical \([TP]_{\text{crit}}\) and at a subcritical \([T_{\text{super}}P_{\text{sub}}]\) condition. It can be seen that the imbibition of brine that follows the loss of free CO₂ to the aqueous phase occurred via both snap-off and piston-like mechanisms. The amount of imbibition was obviously proportional to the fraction of free CO₂ that could be dissolved. At \(T_{\text{super}}P_{\text{sub}}\) all of the free gas was dissolved before the brine became fully saturated, whilst at \((TP)_{\text{crit}}\) only 38% of the injected gas (representing 0.057PV) was dissolved before brine reached saturation point. Note that even when the larger part of the injected gas is not dissolvable (as is the case at \((TP)_{\text{crit}}\)) snap-off imbibition mechanisms during the earlier dissolution phase means that the remaining free gas is likely to be in a disconnected and less mobile form.
4.3.2.1.1 Impact of Network Architecture on Dissolution Rate

The network architecture evidently had an impact on the dissolution rate which cannot be fully accounted for by the differences in mean pore radii alone, as Figure 4-35 shows (for the supercritical condition \((TP)_{super}\)). It is striking that the network with the largest mean pore radius (and hence the largest diffusion surface area) did not exhibit the fastest dissolution rate (Figure 4-35). In constructing the networks used in these sensitivities, a constant pore length was used throughout even as the mean network pore radius was varied. The volume of each pore was also assumed to be equal to the volume of a simple cylinder described by the pore diameter (2x pore radius) and the pore length. These two assumptions have the combined implication that both the pore aspect ratio (radius/pore length) and medium porosity varied from one network to the other. The networks also possess different PSD variances \((\sigma^2=6.75)\) for \(R_{mean}=5.5\mu m\) and \(R_{mean}=35.5\mu m\); \(\sigma^2=200\) for \(R_{mean}=75.5\mu m\); and \(\sigma^2=816.75\) for \(R_{mean}=250.5\mu m\) and this explains the non-monotonic trend between \(R_{mean}\) and dissolution rate – either presented in terms of gas saturation profile with time (as in Figure 4-35-i) or in terms of brine concentration profile with time (as in Figure 4-35-ii). A single PSD variance was not used for all networks to avoid pathologically unrealistic network architectures given the \(R_{mean}\) range that was investigated. For example, the \(R_{min}\) and \(R_{max}\) for the network with an \(R_{mean}\) of 250.5\(\mu m\) would be 246\(\mu m\) and 255\(\mu m\), respectively, if its PSD variance were 6.75 as in the network with an \(R_{mean}\) of 5.5\(\mu m\).

4.3.2.2 Results of continuous injection simulations

We now present results from simulations involving continuous gas injection, and which account for gas dissolution from the outset. Figure 4-36 compares CO\(_2\) critical saturations \((S_{gc}^{CO_2})\) and CH\(_4\) critical saturations \((S_{gc}^{CH_4})\) as functions of network \(R_{mean}\) at the five temperature and pressure conditions. Except at condition \(T_{sub}P_{super}\), where \(S_{gc}^{CO_2}\) was consistently greater than \(S_{gc}^{CH_4}\) on account of the relatively lower CO\(_2\) IFT/\(\Delta\rho\) ratio (see Figure 4-26e), Figure 4-36 suggests CO\(_2\) and CH\(_4\) exhibited very similar evolution behaviours at the other four conditions. This is a misleading picture and it reveals the limitation of characterising a flow process by a single data point alone (in this case \(S_{gc}\)). Figure 4-37 shows a diversity of important differences between CO\(_2\) and CH4 evolution in equivalent networks, driven by variations in IFT/\(\Delta\rho\) ratio and solubility as a result of varying temperature and pressure conditions.
For reasons which have been discussed in the previous section, the impact of dissolution at pressures above the critical pressure of CO$_2$ (7.38MPa) i.e. at the conditions ($TP$)$_{crb}$, ($TP$)$_{super}$, and $T_{sub}P_{super}$ (Figure 4-37[a, c, e]), was very small. At high pressures, CO$_2$ and CH$_4$ saturations tend to build-up at the same rate, but sufficient discrepancies may exist between CO$_2$ and CH$_4$ IFT/$\Delta\rho$ (or Bond number) ratios (see Figure 4-26e) at these conditions for CO$_2$ and CH$_4$ to exhibit entirely different flow regimes in equivalent networks (Figure 4-37a-iii, 4-37c-iii, 4-37c-iv). For pressures below CO$_2$ critical pressure – conditions ($TP$)$_{sub}$ and $T_{super}P_{sub}$ (Figure 4-37[b, d]) – CO$_2$ dissolution played a more significant role especially as $R_{mean}$ increased. The high dissolvability of CO$_2$ at these conditions means that CO$_2$ build-up can be considerably delayed compared to CH$_4$ build-up in equivalent networks (Figure 4-37[b-iv, d-iv]), and although CO$_2$ saturation may eventually rise to the levels reached by CH$_4$ (as brine becomes saturated), subsequent CO$_2$ flow behaviour is invariably affected by the earlier changes to CO$_2$ cluster structure and configuration due to dissolution and the accompanying water imbibition.

4.3.3 Implications of the results

At four out of the five temperature and pressure conditions considered, $S_{gc}^{CH_4}$ may be substituted for $S_{gc}^{CO_2}$ with relatively little loss of accuracy. However, the apparent closeness of $S_{gc}^{CH_4}$ and $S_{gc}^{CO_2}$ at these conditions masks important differences between CO$_2$ and CH$_4$ evolution behaviours in equivalent networks which only the examination of the full saturation profiles can reveal – this makes the use of $S_{gc}$ as a screening tool highly limiting. Moreover, both $S_{gc}^{CH_4}$ and $S_{gc}^{CO_2}$ vary from one temperature and pressure condition to another to such a degree that the substitution of $S_{gc}^{CH_4}$ or $S_{gc}^{CO_2}$ derived at one temperature and pressure condition for use at a sufficiently different condition would be highly erroneous. Figure 4-38 shows that, regardless of the type of gas injected, $S_{gc}$ could vary by more than 100% through temperature and pressure changes alone. This effect is enhanced as the mean pore radius increases – i.e. as the influence of gravity increases.

The suppressed impact of solubility at high pressures (>10.34MPa), even at very low capillary numbers used here, suggests that CO$_2$ flow experiments can produce reliable
results without the need to pre-equilibrate the brine (prior saturation of the brine with CO₂) before experiments.

Although gas solubility does not appear to play a major role in determining $S_{gc}$ generally, we should note that the impact of solubility on the gravity-driven regimes considered here could have been affected by the 2D nature of the networks and their relatively small size. However, even for large 3D systems, over the range of flow velocities likely to be encountered during the injection and subsequent migration of free supercritical CO₂, the rate of loss of free CO₂ to the aqueous brine via molecular diffusion might be too slow to cause large perturbations of the migration pattern of the free CO₂ stream. Dissolution effects may only become significant after all free CO₂ flow has practically ceased.

Table 4-4: Detailed Fluid Properties near CO₂ critical point (T= 35°C, P= 10.34MPa).

<table>
<thead>
<tr>
<th>Fluid Properties</th>
<th>Fluid or Fluid Combinations</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>IFT, [mN/m]</td>
<td>CO₂-H₂O</td>
<td>27.6</td>
</tr>
<tr>
<td></td>
<td>CH₄-H₂O</td>
<td>62.1</td>
</tr>
<tr>
<td>Density, [kg/m³]</td>
<td>CO₂</td>
<td>695.2</td>
</tr>
<tr>
<td></td>
<td>CH₄</td>
<td>78.052</td>
</tr>
<tr>
<td>Δρ, [kg/m³]</td>
<td>CO₂-H₂O</td>
<td>304.8</td>
</tr>
<tr>
<td></td>
<td>CH₄-H₂O</td>
<td>921.948</td>
</tr>
<tr>
<td>$Bo^{CO₂-H₂O}/Bo^{CH₄-H₂O}$</td>
<td></td>
<td>0.74</td>
</tr>
<tr>
<td>Solubility, [kg/m³]</td>
<td>CO₂</td>
<td>45.93</td>
</tr>
<tr>
<td></td>
<td>CH₄</td>
<td>0.872</td>
</tr>
<tr>
<td>Compressibility factor</td>
<td>CO₂</td>
<td>0.252</td>
</tr>
<tr>
<td></td>
<td>CH₄</td>
<td>0.871</td>
</tr>
<tr>
<td>Viscosity, PaS</td>
<td>CO₂</td>
<td>5.82E-05</td>
</tr>
<tr>
<td></td>
<td>CH₄</td>
<td>1.41E-05</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$R_{mean}$, μm</th>
<th>5.5</th>
<th>35.5</th>
<th>75.5</th>
<th>250.5</th>
</tr>
</thead>
<tbody>
<tr>
<td>$σ²$</td>
<td>6.75</td>
<td>6.75</td>
<td>200</td>
<td>3300</td>
</tr>
</tbody>
</table>

Figure 4-20: Comparison of (a) CH₄-H₂O and (b) CO₂-H₂O regime transitions with variation in $R_{mean}$ at $(TP)_{crit}$. Gas dissolution deactivated.
Figure 4.21: Comparison of Bo profiles of $\text{CO}_2$ and $\text{CH}_4$ as functions of PVI, for network $R_{\text{mean}}$ of (i) 5 $\mu$m (ii) 35 $\mu$m (iii) 75 $\mu$m, and (iv) 250 $\mu$m, with gas dissolution disabled.

Figure 4.22: Comparison of $S_g$ profiles of $\text{CO}_2$ and $\text{CH}_4$ as functions of PVI, for network $R_{\text{mean}}$ of (i) 5 $\mu$m (ii) 35 $\mu$m (iii) 75 $\mu$m, and (iv) 250 $\mu$m, with gas dissolution deactivated.
Figure 4-23: The impact of dissolution on CH$_4$ saturation profiles at different network $R_{\text{mean}}$ of (i) 5 µm (ii) 35 µm (iii) 75 µm, and (iv) 250 µm. D denotes active dissolution.

Figure 4-24: The impact of dissolution on CO$_2$ saturation profiles at different network $R_{\text{mean}}$ of (i) 5 µm (ii) 35 µm (iii) 75 µm, and (iv) 250 µm. D denotes active dissolution.
Figure 4-25: Comparison of $S_g$ profiles of CO$_2$ and CH$_4$ as functions of PVI, for network $R_{\text{mean}}$ of (i) 5 µm (ii) 35 µm (iii) 75 µm, and (iv) 250 µm, with gas dissolution activated (to be compared to Figure 4-24).

Figure 4-26: The impact of CO$_2$ dissolution on flow. (a) No dissolution, (b) CO$_2$ dissolving.
Figure 4-27: Comparison of fluid properties of CH$_4$-H$_2$O and CO$_2$-H$_2$O at the five temperature and pressure conditions. CO$_2$-H$_2$O IFT was averaged from: Bachu and Bennion (2009), Nielsen et al (2011), Espinoza, and Santamarina (2010), and Li et al (2012), while CH$_4$-H$_2$O IFT was averaged from: Hough et al (1951), Jennings et al (1971), and Shariat et al (2012) at the temperature and pressure conditions. Viscosity and density data obtained from NIST (National Institute of Standards and Technology) database. The Bond number (IFT/Δρ) ratio chart (e) is added here for completeness.

Figure 4-28: Comparison of $S_g$ profiles of (a) CH$_4$ and (b) CO$_2$ as functions of shut in time at five temperature and pressure conditions.
Figure 4-29: CO₂ density as a function of temperature and pressure, using Redlich-Kwong EOS.

Figure 4-30: CO₂ solubility in pure H₂O as a function of temperature and pressure, using the model of Duan et al (2006).

Figure 4-31: CO₂ dissolvability in pure H₂O as a function of temperature and pressure.
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Figure 4-32: Comparison of the correlation of total CO₂ dissolved (as a fraction of network PV) to (a) the CO₂-H₂O solubility (Kg_{CO₂}/Kg_{H₂O}) and (b) the CO₂-H₂O dissolvability index (m³_{CO₂}/m³_{H₂O}) at the five temperature and pressure conditions and for a network R_{mean} of 75µm.

Figure 4-33: Comparison of average aqueous CO₂ concentration profiles as a function of shut in time at five temperature and pressure conditions for a network R_{mean} of 75µm using (a) a semi-log plot and (b) a Cartesian plot. The empty lines represent equilibrium lines.
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Figure 4-34: The variation of free CO\textsubscript{2} saturation and dissolved CO\textsubscript{2} concentration distributions with shut in time at (a) a supercritical condition (TP\textsubscript{crit}) and, (b) a subcritical condition T\textsubscript{super}P\textsubscript{sub}.

Figure 4-35: The impact of network architecture on CO\textsubscript{2} dissolution rate presented in terms of (i) free CO\textsubscript{2} saturation profile with time, and (ii) Aqueous CO\textsubscript{2} concentration (in brine) with time, at supercritical temperature and pressure (TP\textsubscript{super}; T=100°C, P=3500psi). Dotted line is the equilibrium line.
Figure 4-36: Comparison of $S_{gc}$ profiles of CH$_4$ and CO$_2$ as functions of network R$_{mean}$, with gas dissolution activated at (a) (TP)$_{crit}$; (b) (TP)$_{sub}$; (c) $T_{sub}P_{super}$; (d) $T_{super}P_{sub}$; (e) (TP)$_{super}$. 
Figure 4-37: Comparison of CO$_2$ and CH$_4$ $S_g$ profiles as functions of PVi, for network $R_{mean}$ of (i) 5 µm (ii) 35 µm (iii) 75 µm, and (iv) 250 µm, at (a) $(TP)_{crit}$; (b) $(TP)_{sub}$; (c) $T_{sub}P_{super}$; (d) $T_{super}P_{sub}$; (e) $(TP)_{super}$, with gas dissolution activated. Rows correspond to changing mean pore radius while columns correspond to changing temperature and pressure conditions.
Figure 4-38: The impact of T and P upon (A) CH₄ and (B) CO₂ S_{gc} as functions of model R_{mean} for (i) Inert and (ii) Reactive flows

4.4 Summary and Conclusions

The dynamic pore network model of multiphase flow in porous media described in Chapter 3 has been used to perform a parametric sensitivity study of gas injection regimes under the influence of capillary and gravity forces. A regime classification scheme was developed and subsequently used to analyse the characteristic flow behaviors (including coupled effects of non-equilibrium mass transfer) of CO₂-H₂O and CH₄-H₂O binary fluid systems at different temperature and pressure conditions. Important conclusions are highlighted below.

- A selective parametric sensitivity of key variables that define the local Bond number – R_{mean}, IFT, Δρ, and system height – revealed that gravity-driven regimes can be grouped into two broad classes: (a) stable [Bo < 1] and (b) unstable (migratory) flow [Bo ≥ 1]. Changes in the critical gas saturation (S_{gc}) in response to the variation in any one of the parameters of the Bo was found to depend strongly on the prevailing regime and the topological properties of the network.
• Simulation results predict that the change in the strength of the gravitational gradient from a core scale to a gridblock scale could lead to fundamental changes in flow regimes which have so far been neglected in current upscaling procedures. Particular attention has been drawn to the effects of these scale-dependent transitions on gas breakthrough saturation, the residual gas saturation distribution and the gas-liquid contact area available for diffusion. Success in accurately quantifying these effects would depend to a large extent on the precision with which IFTs, fluid densities and the pore size distributions are measured.

• The PSD variance and pore connectivity were found to have a very strong influence upon gas evolution pattern and on $S_{gc}$ – even though both variables are unaccounted for in standard Bond number definitions.

• Comparisons of CH$_4$ and CO$_2$ regimes injected into equivalent networks at five different combinations of temperatures and pressures have been presented. Results showed that CH$_4$-H$_2$O systems exhibited higher Bond numbers and were therefore more gravity biased than CO$_2$-H$_2$O systems. Gas dissolution had a complicated effect on flow regime - dissolution shrinks gas structures but may also alter their topology, leading to the exploration of new flow pathways which might result in an increase or a decrease in breakthrough saturation. The impact of dissolution was most pronounced at low system pressures (≤0.69MPa). The higher solubility of CO$_2$ in water compared to CH$_4$ tended to reduce differences between $S_{gc}^{CH_4}$ and $S_{gc}^{CO_2}$ (breakthrough saturations of CH$_4$ and CO$_2$, respectively) which might be caused by the greater gravity bias of CH$_4$ (except in the case where the superior net gravity forces on CH$_4$ flow led to a regime transition from stable to migratory).

• In four out of the five temperature and pressure combinations considered, differences between $S_{gc}^{CH_4}$ and $S_{gc}^{CO_2}$ were not significant. However, at a temperature of 25°C and a pressure of 24.13MPa, $S_{gc}^{CO_2}$ was more than double $S_{gc}^{CH_4}$ – essentially because CO$_2$ flow was characterized by much lower Bond numbers than CH$_4$ flow under this particular condition. For temperatures and pressures above the CO$_2$ critical point, CO$_2$ and CH$_4$ flow regimes in brine displayed generally similar characteristics, suggesting
that flow coefficients (e.g. relative permeability) of CH₄ and CO₂ in brine could be used interchangeably in continuum type simulators with effectively the same results. For conditions with temperatures below the critical point of CO₂ but pressures above the critical point of CO₂, CH₄ flow properties cannot be substituted for CO₂ properties without considerable adjustments.

- Gas solubility in brine is a poor guide to predicting the volumetric fraction of the injected gas (CO₂ or CH₄) that can be dissolved as pressure changes. Solubility generally increases with pressure but it was also found that the fraction of injected gas that can dissolve decreases with increase in pressure since gas density also increases. A dissolvability index is proposed to help better correlate the dissolution capacity of brine to CO₂ as system pressure changes.

- The CO₂ solubility trapping capacity of brine at conditions typical of candidate aquifers for CO₂ storage is probably limited to about 0.06PV of aquifer equivalence of total injected CO₂ i.e. brine may not dissolve CO₂ greater than 1/20th PV of aquifer before complete saturation occurs. However, our simulations show that if 38% of total injected CO₂ can be dissolved then snap-off imbibition mechanisms during the dissolution can help to reduce the connectivity of the undissolved CO₂ and render it less mobile.

- The salient features of the gas saturation patterns resulting from two injection experiments by Dumoré (1970) and Geistlinger et al (2006) were successfully reproduced, giving additional confidence in our modeling approach.

Low injection rates were used in the simulations throughout this chapter in order to negate the effect of viscous forces on flow. In the next chapter, higher injection rates will be considered, allowing us to examine the impact of the coupled effects of capillary, gravity, and viscous forces on regime transitions.