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**Non-Uniqueness in Capillary Pressure-Saturation-Relative Permeability
Relationships for Two-Phase Flow in Porous Media:
Interplay between Intensity and Distribution of Random Micro-Heterogeneities**

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Non-Uniqueness in Capillary Pressure-Saturation-Relative Permeability Relationships for Two-Phase Flow in Porous Media: Interplay between Intensity and Distribution of Random Micro-Heterogeneities

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ABSTRACT

The two-phase flow behaviour in porous media is determined on the basis of capillary pressure-saturation-relative permeability relationships (P^c - S - K_r). These relationships are highly non-linear and obtained by laboratory experiments on porous samples, typically around 10-12cm in length. It is normally assumed that these samples are homogenous; however it is well known that this is in fact not the case and that even at this scale micro-scale heterogeneities exist. Two-phase flow experiments on soils with different properties (e.g., particle and pore size distribution, permeabilities, etc) result in different P^c - S relationships. These cause non-uniqueness in the P^c - S curves. Recent work has shown that the presence of these micro-heterogeneities has a significant effect on the measured P^c - S - K_r relationships and they also cause non-uniqueness in these relationships. In the previous work in this area, the micro-heterogeneity effects on the P^c - S - K_r relationships have been analysed in a number of contexts, e.g., uniformly distributed heterogeneities (simplified cases), various binary sand combinations, hydraulic parameters (e.g., entry pressure, permeability), boundary conditions, etc. There is also some evidence that the intensity and distribution of the micro-heterogeneities affect the P^c - S - K_r relationships. In the present work we use numerical simulations to investigate further the nature of these effects, in particular how the interplay between the intensity and random distribution of micro-heterogeneities affects the P^c - S - K_r relationships. Seven randomly heterogeneous patterns have been defined. These domains represent coarse sand media with fine sand blocks embedded in them. The domain size has been chosen (12 cm $\times$ 12 cm) so that it represents a typical laboratory scale device. The results of the simulations show that it is particularly important to take into account both the intensity and distribution of heterogeneity when determining the effective P^c - S - K_r relationships of a sample. This is in contrast to the results of domains with uniformly distributed heterogeneities. We have found that in general if the intensity of heterogeneity is high; the irreducible wetting phase saturation (S_{iw}) of the sample is also high. The direction of flow and the orientation of the samples also have significant effects. For example, the injection of an immiscible phase from the top (vertically downward) of water saturated porous domain leads to a lower S_{iw} than injecting on horizontal plane. On the other hand, injection from the bottom (vertically upwards) leads to a higher S_{iw} . As expected, the distribution of heterogeneity has a significant effect on the saturation distribution and the shape of the P^c - S - K_r curves. However, we show that if the heterogeneities are distributed in such a way that they are closer to the boundary of injection, the irreducible wetting phase saturation is higher.

KEYWORDS: Non-aqueous phase liquid (NAPL), porous media, micro-heterogeneity, multi-phase flow, upscaling, P^c - S - K_r relationships

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1. Introduction

Non-aqueous phase liquids (NAPLs) have been widely manufactured and/or used in various chemical process industries (CPI). These NAPLs include hazardous organic compounds, such as solvents, chlorinated and fluorinated hydrocarbons, transformer oils and others. Accidental spills and leakage of these chemicals from various sources (e.g., underground storage tanks) in the hydro-environmental systems pose a serious long-term threat to the environment. This is primarily because (a) the NAPLs have very low solubility in water and hence, remains as a separate phase (b) chemical decomposition of NAPLs is extremely slow and they may take hundreds of years for natural disappearance (c) they are highly mobile and can contaminate below the water table which is generally the source of drinking water. While the NAPL contamination continues to persist as a problem in the industrialized nations, this type of contamination has received little attention in the developing world (www.iwahq.org.uk/template.cfm?name=sg49). As lesser developed nations continue their progress towards industrialization, it is feared that the similar problems that have been encountered in the USA and Europe in the last couple of decades will appear in the developing world. To predict the flow of NAPL in the subsurface, assess the amount of entrapped NAPL volume and to design accurate subsurface remediation techniques, it is therefore necessary to enhance our understanding and improve the description of the immiscible two-phase flow behaviour (i.e., water and NAPL in this case) in porous media.

The density of the NAPLs has a decisive influence on their flow behaviour. Therefore, the NAPLs are grouped into two classes: (i) LNAPLs (light NAPLs) which have a density less than that of water and, hence float and mostly remain on top of the water table and, (ii) DNAPLs (dense NAPLs) which are denser than water and, thus can penetrate the water table and move deeper into the aquifer system. The viscosities of the NAPLs also play a role in determining the flow behaviour. In general, LNAPLs have a viscosity higher than that of water. On the other hand, DNAPLs are generally less viscous than water.

However, the two-phase flow behaviour in the subsurface is a very complex process and is governed by a number of material dependent parameters (discussed below) and the relationships between capillary pressure (P^c), saturation (S) and relative permeability (K_r) (Collins, 1961; Bear, 1972; Helmig, 1997). The P^c - S relationships quantify the level of saturation (water content or level of fluid hold up by volume) as a function of capillary pressure in porous media. On the other hand, the K_r - S relationships quantify the relative permeability of the porous domain to a particular fluid phase (e.g., NAPL) at different fluid saturation values. The effects of all fluid and material properties are then lumped into the non-linear P^c - S - K_r curves for multiphase flow in a porous domain. Two-phase flow experiments on soils with different properties (e.g., particle and pore size distribution, permeabilities, etc) result in different P^c - S - K_r curves. These cause non-uniqueness in the P^c - S - K_r curves.

The P^c - S - K_r relationships are also affected by the flow conditions (steady or dynamic) within, and at the boundary of, the domain. For a given flow type, these relationships are again influenced by the interplay of a number of other factors, e.g., initial water saturation in the domain; wettability of the solid grains to

particular fluid phase; contact angles between the fluid phases and solid grains; pore and particle size distribution; fluid properties (e.g., viscosity, density); interfacial tension; gravity forces; types of heterogeneities; domains size; hysteresis, etc (Palosaari and Cornish, 1975; Mitlin et al., 1998; Oostrom, M. and Lenhard, R J, 1998; Hassanizadeh et al., 2002; Tsakiroglou et al., 2003; Das et. al., 2004; Ishakoglu and Filiz Baytas, 2005; Hilfer, 2006). Because of a complex interplay between these factors, it is generally not possible to measure unique P^c - S - K_r relationships for a porous sample. This introduces difficulties in using the measured P^c - S - K_r relationships to practical problems. Therefore, there have been a number of attempts to characterize how the variations in the above factors cause non-uniqueness in these relationships in an attempt to generalize their effects as far as possible (e.g., Ataie-Ashtiani *et. al.*, 2002, 2003; Das et. al., 2004). In other cases, attempts have been made to improve the predictability of multiphase flow models by developing various types of theories and scaling rules (e.g., Busby et al., 1995; Desbarats, AJ, 1995; Bradford, and Leij, 1996; Ataie-Ashtiani et. al., 2001).

In general, the P^c - S - K_r relationships are obtained by laboratory experiments on soil samples, typically around 10-12cm in length (core scale). At this scale, it is generally assumed that the domain is a homogeneous medium. However even at that scale some form of micro-scale heterogeneities exists. These heterogeneities are much bigger than the pore-scales but relatively small as compared to the scales of the laboratory device. Therefore, the micro-heterogeneities are often viewed as sub-sample scale heterogeneities and, as expected, may vary in size, shape and distribution from one domain to another. These variations are known to cause non-uniqueness in the P^c - S - K_r relationships, as confirmed by many experimental and numerical studies (Ataie-Ashtiani *et. al.*, 2001, 2002, 2003; Oung & Bezuijen, 2003; Das et. al., 2004). Non-uniqueness in the P^c - S - K_r relationships also arises because the heterogeneities have distinctly different multi-phase flow properties (i.e., P^c - S - K_r relationships) from that of the surrounding media. As a result, they play a crucial role in determining the extent and temporal aspects of the spreading behaviour of NAPL, which affect the amount and distribution of entrapped fluid in the domain (residual saturation). The micro-heterogeneities are often sources of localised pools of NAPLs and cause rise of local NAPL concentration to dangerous values. It is therefore of great significance that the determination of P^c - S - K_r relationships at the laboratory scale for multi-phase flow in porous media account for the presence of micro-heterogeneities in the samples.

However, characterizing the micro-heterogeneity effects on P^c - S - K_r relationships is not straight forward because of the small size of the heterogeneities. Laboratory experiments, which allow inclusion and control of random micro-scale heterogeneities, are difficult to design. Experiments may be conducted with carefully controlled uniform heterogeneities (e.g., Kueper and Abbot, 1989; Dawe et. al., 1992); however, the P^c - S - K_r relationships for uniformly distributed heterogeneities cannot be used directly for domains with random heterogeneity distribution because of the very different shapes of the P^c - S - K_r curves (Das et al., 2004). Experiments may be conducted with real samples from the subsurface but the intensity and the distribution of micro-heterogeneities in these domains cannot be controlled, although perhaps characterized using expensive non-invasive techniques. It is therefore not trivial to correlate the variation and intensity of the random micro-heterogeneities to the P^c - S - K_r relationships. Consequently, the preferred option for characterizing the

micro-heterogeneity effects on P^c - S - K_r relationships has been the use of numerical modelling (Ataie-Ashtiani *et. al.*, 2001, 2002; Das *et. al.*, 2004; Manthey *et al.*, 2005; Selim *et. al.*, 2005). Most authors in this area have experimented on two-dimensional domains orientated in the horizontal plane so as to negate the effects of gravity and thereby synthesise the micro-heterogeneity effects more conclusively (e.g., Ataie-Ashtiani *et. al.*, 2001, 2002; Das *et. al.*, 2004). On the other hand, other authors (e.g., Ataie-Ashtiani *et. al.*, 2003, Oung & Bezuijen, 2003) combine experimental and numerical modelling to study two-phase flow behaviour in 1D where the effects of the other two dimensions on the flow behaviour are ignored. The majority of these works have been conducted on domains with simplistic, evenly distributed heterogeneity patterns instead of more realistic randomly distributed heterogeneities.

Das *et. al.* (2004) have explored how the variations in the nature, amount and distribution of micro-heterogeneities affect P^c - K_r - S relationships for dense non-aqueous phase liquid (DNAPL) and water flow. Several results obtained by Das *et. al.* carry particular relevance to this paper. It has been observed that coarse sand (more permeable) micro-heterogeneities embedded in fine sand (less permeable) have little difference with the P^c - K_r - S curves of background fine sand. However, if fine sand blocks of micro-heterogeneities are embedded in coarse sand, then the P^c - K_r - S relationships are significantly different from those of both coarse and fine sand. In particular, water becomes entrapped in the fine sand heterogeneities and the average irreducible wetting-phase saturation (S_{iw}) in the heterogeneous domain becomes significantly higher than those of homogeneous coarse and fine sand. Das *et. al.* have also noted that the intensity of the micro-heterogeneities (<0.25) has little effect on the P^c - K_r - S curves for the same binary sand combination, although, in general, the higher is the intensity of heterogeneity the greater is the S_{iw} . However, it has been indicated that the distribution of the heterogeneities (i.e., random or uniform) has a significant effect on the two-phase flow behaviour. In other words, if the micro-heterogeneities are imbedded in a random fashion, then for the same binary sand combination, the P^c - K_r - S curves become very different from those for the homogeneous coarse and fine sand and, the medium with uniformly distributed micro-heterogeneities (Das *et. al.*, 2004).

The present paper is largely a follow up from the above conclusions. We seek to extend these conclusions to provide further understanding on how the random micro-heterogeneities affect the two-phase flow behaviour in porous media. In particular, we aim to quantify how the interplay between the intensity and random distribution of micro-heterogeneity within a sample affect the upscaled capillary pressure-saturation-relative permeability (P^c - S - K_r) relationships. We also investigate the effects of flow directions (e.g., along/against and without gravity) on P^c - S - K_r relationships for random heterogeneity patterns, attempting to generalise their effects as far as possible. A number of authors (Ataie-Ashtiani *et. al.*, 2001; 2002; Das *et. al.*, 2004; Selim *et. al.*, 2005) have introduced the sub-sample scale heterogeneities in the domain by defining different size of the heterogeneous blocks. In this work, we vary the size, distribution and amounts of heterogeneities. These enable us to correlate both the intensity and the distribution of heterogeneities to P^c - S - K_r relationships.

155 We also compare P^c -S relationships for representative patterns with uniformly and randomly distributed
 156 micro-heterogeneities.

157

158 Our solution procedure starts at the sub-sample scale where the conservation equations for momentum and
 159 mass are assumed known. The P^c -S- K_r relationships for these blocks are specified using standard empirical
 160 relationships. Subsequently, simulations are carried out to derive the effective P^c -S- K_r curves at the larger
 161 scale (core scale) that take into account the complexity of the medium and the sub-sample scale processes.
 162 The governing equations used in this work are discussed in the next section. In this paper, we present results
 163 based on our steady state simulations for two-phase flow in 2D domains.

164

165 **2. Description of numerical simulations for two-phase flow in porous media**

166 Before discussing the governing model equations, it is also worthwhile to conceptualise the two-phase flow
 167 behaviour for the purpose of this work. We specifically focus on the flow of two immiscible fluid phases in
 168 porous media: (a) DNAPL, called as the non-wetting phase and (b) water, called as the wetting phase. To
 169 begin with the porous domain (homogeneous or heterogeneous) is defined to be fully saturated with water.
 170 The non-wetting phase is then forced to infiltrate, which drains water out of the domain (*drainage*
 171 *experiments*). It is known that the most NAPLs are slightly soluble in water over a long period of time
 172 (Mayer and Hassanizadeh, 2005). However, for the purpose of this paper (shorter time durations) the NAPL
 173 dissolution in water is assumed to have no effect on the two-phase flow behaviour.

174

175 **2.1 Governing model equations**

176 Modelling the two-phase flow processes involves the solution of the conservation laws for mass and
 177 momentum supplemented by appropriate constitutive formulae for P^c -S- K_r relationships. The following form
 178 of Darcy's equation is used as the conservation equation of motion for non-wetting and wetting phases:

$$179 \quad \mathbf{q}_\gamma + \frac{K_r \mathbf{k}}{\mu_\gamma} \cdot \nabla P_\gamma = 0 \quad \text{for} \quad \gamma \equiv w, nw \quad (1)$$

180 In equation (1), the subscripts 'w' and 'nw' are the wetting (water) and non-wetting (DNAPL) fluid phases,
 181 respectively; $\mathbf{q}$ is fluid flow velocity [LT^{-1}], K_r is relative permeability [-], $\mathbf{k}$ is intrinsic permeability tensor of
 182 porous media [L^2], μ is fluid viscosity [$ML^{-1}T^{-1}$] and P is average pore pressure [$ML^{-1}T^{-2}$] of fluid.

183

184 The conservation of mass in the two-phase flow system is governed by the following form of continuity
 185 equation,

$$186 \quad \frac{\partial}{\partial t} (\phi \rho_\gamma S_\gamma) + \nabla \cdot (\rho_\gamma \mathbf{q}_\gamma) = 0 \quad \text{for} \quad \gamma \equiv w, nw \quad (2)$$

187 where ϕ is the porosity of the medium [-], ρ is fluid density [ML^{-3}], and S is the average fluid saturation [-] in
 188 the porous medium.

189

190 The capillary pressure-saturation (P^c - S) relationships are given by the Brooks-Corey formulations (Brooks
191 and Corey, 1964),

$$192 \quad S_{ew} = \left(\frac{P^c}{P^d} \right)^{-\lambda} \quad \text{for } P^c \geq P^d \quad (3)$$

$$193 \quad S_{ew} = 1 \quad \text{for } P^c \leq P^d \quad (4)$$

$$194 \quad S_{ew} = \left(\frac{S_w - S_{rw}}{1 - S_{rw}} \right), \quad 0 \leq S_{ew} \leq 1 \quad (5)$$

195 where S_{ew} is the effective saturation [-] of the wetting phase, P^d is entry pressure [$\text{ML}^{-1}\text{T}^{-2}$] of the medium, λ
196 is a pore size distribution index [-] and S_{rw} is irreducible wetting phase saturation [-].

197

198 In order to calculate the relative permeabilities of the porous samples to the wetting (K_{rw}) and non-wetting
199 phase (K_{rnw}), the empirical Brooks-Corey-Burdine formulae (Brooks and Corey, 1964) are used,

$$200 \quad K_{rw} = S_{ew}^{(2+\lambda)/\lambda} \quad (6)$$

$$201 \quad K_{rnw} = (1 - S_{ew})^2 (1 - S_{ew}^{(2+\lambda)/\lambda}) \quad (7)$$

202

203 The governing equations are solved using the code STOMP (Subsurface Transport Over Multiple Phases).
204 This simulator has been specifically designed for the use in the modelling of multiphase flow in porous
205 media and has been used for a wide variety of research (White *et. al.*, 1995; Oostrom *et. al.*, 1997; Oostrom
206 and Lenhard, 1998; Ataie-Ashtiani *et. al.*, 2001, 2002; Oostrom and White, 2003; Das *et al.*, 2004; Wipfler *et.*
207 *al.*, 2004). The spatial discretisation of the governing equations in STOMP is done based on the standard
208 finite volume method, FVM (Pantakar, 1980; Versteeg and Malalasekera, 1995). The non-linear equations
209 derived through the FVM discretisation are reduced to linear equations using the Newton-Raphson iterative
210 method for multiple variables. The temporal discretisation is based on the well-known implicit time stepping
211 method. The validity and utility of the code for the kind of problems in this work have been shown by
212 previous studies (Ataie-Ashtiani *et. al.*, 2001; Das *et. al.*, 2004; Wipfler *et. al.*, 2004) and are not discussed in
213 this paper.

214

215 **2.2. Characteristics of porous media and fluids**

216 In this section, we describe the heterogeneous porous domains with reference to: (i) the intensity of
217 heterogeneity, ω [-] which is defined as the ratio of the total area of micro-heterogeneities to the total
218 sample area, and (ii) the distribution of heterogeneity. Seven different random micro-heterogeneity patterns
219 have been defined where each pattern is a two-dimensional (2D) block of $12\text{cm} \times 12\text{cm}$ (Figure 1). The
220 patterns are composed of two different types of sand. While the heterogeneities are made up of fine sand
221 (less intrinsic permeability), the background medium is a coarse sand (high intrinsic permeability). The
222 heterogeneous patterns are named Random 1 (R1) to Random 7 (R7). As the name implies these domains
223 contain an essentially random distribution of micro-scale heterogeneities. However, we control the amounts
224 of imbedded porous media in order to determine the intensity of heterogeneity (ω) of each pattern. The

parameter ω for each pattern is shown in Table 1. The properties of the porous media including the parameters of Brooks-Corey-Burdine equations (Equations 3-7) are shown in Table 2. No correlation is assumed between the porous media properties. For this paper, we choose a common DNAPL, namely PCE (tetrachloroethylene). The fluid properties used in our simulation are also shown in Table 2.

2.3 Initial and boundary conditions for numerical simulations

As mentioned before, the simulations have been carried out on 2D domains of coarse sand with micro-heterogeneous blocks of fine sand embedded in them (Figure 1). Initially, the sample is fully saturated with water. A boundary of injection for DNAPL is then chosen, which is either from Top or Bottom of the sample for vertically aligned sample or from North side of the block for horizontally aligned domain. It is also worth defining here the convention used to specify the orientation of a sample. North, South, East and West are used to represent the positive and negative x- and y-axes, respectively, whilst the Top and Bottom represent the positive and negative z-axes. Therefore, if an injection in a 2D sample is said to occur from the North it implies that the domains is orientated in the horizontal plane and the DNAPL is injected from North side of the domain. If an injection is said to take place from the Top or Bottom they imply that the domains are orientated on a vertical plane. The boundary conditions are defined such that the two fluid phases enter the domain through the injection face and flow out through the drainage face. The sidewalls are defined to be impermeable and defined as ‘no flow boundaries’ for both phases. In effect, our simulations correspond to the flow through type cells for measuring P^c - S - K_r relationships. In all simulations, a pressure difference (ΔP) of 1000 Pa across the domain is imposed for both wetting and non-wetting phases. The capillary pressure ($P^{nw}-P^w$) is chosen to be the same at both injection and drainage faces. The imposed capillary pressure is varied by changing P^{nw} as described in detail below.

The pressure of the DNAPL is initially zero everywhere in the domain, apart from the boundary of injection where it is equal to the pressure of the wetting phase. This yields a capillary pressure (P^c) equal to zero at the boundary initially. Then, the pressure of the non-wetting phase is gradually increased, whilst the pressure of the wetting phase is kept constant. This leads to an increase in P^c at the boundary of injection. Once P^c reaches the entry pressure (P^d) for the background medium (370 Pa for coarse sand in this case, Table 2), DNAPL and water with different pressure infiltrates into the sample and initial water in the domain is drained out by injected DNAPL. The simulation is carried out until steady-state flow conditions are reached for each P^c , that is, saturation at the numerical grid points does not vary any more. The average saturation of the sand sample at steady state and the imposed P^c provide one point of the P^c - S graph. Next, the imposed capillary pressure is increased incrementally by increasing DNAPL pressure and the simulation is continued until a new steady state is reached. A second point for P^c - S is thus obtained. This procedure is repeated until a P^c of 11000 Pa is reached by which point the sample is deemed to have reached its irreducible wetting phase saturation (S_{iw}). This practically means that no more water can be drained out of the sample regardless of the capillary pressure. At the opposite boundary both the values of DNAPL and water pressure remain constant.

Figure 2 depicts these boundary conditions graphically for the case of a vertically aligned pattern where DNAPL is injected at the top (T) of the domain while water drains out from the bottom (B) of the sample.

2. 4 Convergence of numerical results

The numerical grid used in our simulations involves a uniform non-staggered mesh consisting of 48×48 square cells of size $2.5 \text{ mm} \times 2.5 \text{ mm}$. The convergence of the numerical solution is achieved based on a maximum Newton iteration of either 16 or 32, depending on the complexity of the domain type and a maximum tolerance limit of 10^{-6} . However, the convergence of the results has been also checked through mesh refinement. Ideally, this test should be carried out using a grid with infinitesimal small nodal spacing and comparing the results from the chosen and refined meshes. However, this has been found almost impossible in our case since the patterns (Figure 1) contain random heterogeneities and the computational time increases prohibitively when the normal mesh is refined more than four fold. In this work, the grid size is changed four fold for the convergence test. This is done by keeping the domain size constant ($12 \text{ cm} \times 12 \text{ cm}$) but increasing the number of cells to a desired level. The refined mesh chosen for the test involves 96×96 square cells of size $1.25 \text{ mm} \times 1.25 \text{ mm}$. The results show negligible difference between P^c -S curves from the two meshes. The maximum percentage difference of $\sim 1.5\%$ is found between the P^c -S curves. The test ensures that the numerical results presented in this paper are, in general, independent of the grid used. This also gives confidence that the accuracy of the numerical results is not a major concern in this work.

3. Results and Discussion

3.1 Effects of intensity and random distribution of heterogeneity on effective P^c -S relationships

Although it is accepted that micro-heterogeneity affects two-phase flow properties in porous media, there is a lack of systematic analyses for the relationships between intensity and distribution of heterogeneities and, effective P^c -S- K_r curves. In this section, we seek to explain, in a quantifiable manner, how the intensity and distribution of heterogeneity in porous media cause non-uniqueness in these relationships. For this purpose the P^c -S curves for the seven domains in Figure 1 have been used. The first pattern (R1) has an intensity of heterogeneity (ω) of roughly 0.2. Patterns R2 – R4 have ω equal to approximately 0.1 and patterns R5 – R7 have ω equal to approximately 0.05. However, within a group of similar ω , the distributions of the heterogeneities are very different. Although these layouts are essentially random, care has also been taken to ensure that the distributions contain significant variations. This is because of the relatively small number of patterns used in this work.

We compare the steady state P^c -S curves for the seven domains (R1-R7) with injection from the north side (horizontal plane), top (vertically downward) and bottom (vertically upward) of the domain, respectively. The simulated results are given in Figures 3-5. In each case, the curves created for patterns R1 - R7 form a distinct group depending on the value of ω although they have different heterogeneity distributions. As expected, the domains with lower intensity of heterogeneity have lower irreducible water saturation (S_{iw}), although significantly higher than the S_{iw} in the homogeneous coarse sand (background medium). It is clear

from these results that for a given binary combination of porous medium, the most important factor in determining S_{iw} (hence, the non-uniqueness in P^c - S - K_r curves) is the intensity of heterogeneity.

One may observe that there are some variations in the shape of the P^c - S curves of the patterns with similar intensity of heterogeneity. However these variations can be considered insignificant. For example, consider the domains R5-R7. They have the almost same values of ω and the difference in the S_{iw} value within this group is ~ 0.02 , which is not significant. There is however a noticeable difference in the shape of the curves, particularly when injecting from the north or the top of the patterns. This difference is due to the distribution of the heterogeneities as well as the gravity effects. We consider how gravity causes non-uniqueness in the P^c - S relationships in more detail in the following sections. The K_r - S curves for a sample pattern (R3) in different directions are discussed briefly in section 3.6.

By analysing patterns with uniformly distributed heterogeneities, Ataie-Ashtiani et. al. (2001, 2002) and Das et. al. (2004) have shown that the P^c - S - K_r curves are affected by the entry pressure and pore size distribution of the porous media, among other factors. While these observations are likely to be valid for domains with random heterogeneities, it seems that the intensity and distribution of the heterogeneities are also important factors in determining the shape of P^c - S - K_r curves. However, no attempt is made in this paper to synthesise the sensitivities of these factors and to determine if effects of one parameter mask the effects of another factor on these relationships.

3.2 Effects of random micro-heterogeneities on DNAPL saturation distribution

In this section, we consider DNAPL (PCE) saturation distributions in heterogeneous domains and analyse how the micro-heterogeneities affect the saturation distributions in three representative patterns, R2, R5 and R6 (Figure 1). The pattern R2 is chosen because it has a relatively higher intensity of heterogeneity (~ 0.1) and the heterogeneities are distributed through out the domain. The patterns R5 and R6 are chosen as they have a similar intensity of heterogeneity (~ 0.05) but distinctly different heterogeneity distributions. In R5 and R6, the heterogeneities are generally distributed in the lower and upper half of the sample, respectively.

We compare the saturation plots in R2, R5 and R6 at two different capillary pressures (400 and 11000 Pa) for the case when the PCE is injected vertically downward (along gravity). The capillary pressure value of 400 Pa is selected as this is close to the entry pressure of the background material (370 Pa) and just allows PCE to enter into the domain. The capillary pressure of 11KPa is adopted because the average water saturation at this value reaches close to the irreducible water saturation (S_{iw}).

As shown in Figures 6-7, the differences in the saturation levels in coarse and fine sand are very noticeable, particularly at high capillary pressure. The PCE infiltrates into the background coarse sand when capillary pressure equals the entry pressure of coarse sand. As the capillary pressure is increased, PCE saturation in the domain also increases. However, this increase is mostly observed in the background coarse sand. It is also

338 noticed that at high capillary pressure (e.g., 11 KPa), the saturation is fairly uniform throughout the
339 background sand. But, at low capillary pressure (e.g., 400 Pa), a gradient in the saturation from the top to the
340 bottom of the sample is observed. The saturation is lower at the top and higher at the bottom of the sample
341 (gravity effects on DNAPL).

342

343 A sharp contrast exists in the saturation value across the fine/coarse sand interface, which is because of the
344 large contrast in the intrinsic permeability of the two sands (10^3) and relative permeability values. The fine
345 sand blocks (heterogeneities) in the domain remain at low PCE saturation. This is because the relative
346 permeability of PCE in fine sand blocks is lower than that in the background coarse sand. Therefore, PCE
347 does not infiltrate into these blocks. Similar phenomena have been observed by Ataie-Ashtiani et. al. (2001,
348 2002) and Das et. al. (2004) for horizontal domains where gravity effects are ignored. Our results are
349 consistent with these observations.

350

351 From our simulations, it is also interesting to note that the consideration where the heterogeneities are
352 located (e.g., upper or lower half of the domain) may be important in determining the value of average
353 S_{iw} and, hence, the shape of P^c - S curves. This is particularly so if the two-phase flow behaviour is affected by
354 the gravity. We note that the closer the heterogeneities are to the boundary of DNAPL injection, the higher is
355 the S_{iw} in the domain. As mentioned above, the micro-heterogeneities are generally located closer to the
356 boundary of injection in R6T and away in R5T. Both domains have similar intensity of heterogeneity.
357 However, S_{iw} is higher in R6T than in R5T (Figure 4). Because of their heterogeneity distribution, the
358 opposite pattern occurs when injecting PCE from the bottom of the domain. In this case, the heterogeneities
359 in domain R6B are in general further from the boundary of injection than those in domain R5B. This means
360 that the S_{iw} in R6B is lower than the S_{iw} in R5B when injecting from the bottom of the sample (Figure 5).
361 These discrepancies cause non-uniqueness in $P^c - S$ curves (Figures 4-5). If DNAPL is injected from the
362 north side of the domain, the discrepancy in S_{iw} is less pronounced and consequently there is less variation
363 in the shape of the P^c - S curves (Figure 3).

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365 **3.3 Comparison of effective P^c - S relationships for uniformly and randomly distributed heterogeneity**

366 In this section, we compare the effects of uniformly and randomly distributed micro-heterogeneities
367 on $P^c - S$ curves in an attempt to demonstrate the implications of heterogeneity distributions more
368 conclusively. Figure 8 displays two domains, HP1 and C4, with uniformly distributed micro-heterogeneities.
369 These domains have been adopted from Das et. al. (2004) where the authors have used them for analysing
370 PCE flow with no gravity effects. We compare the $P^c - S$ curves of these blocks with those obtained for
371 PCE flow in the random heterogeneity patterns R1, R2N, R5N and R6N in the horizontal directions (Figure
372 9). The P^c - S curve for the background coarse sand is also included. We ignore the effects of gravity in all
373 domains in order to synthesise the effects of distribution of heterogeneities more conclusively. The

comparison clearly indicates that the inclusion of random micro-heterogeneities results in higher S_{iw} and different capillary pressure curves.

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The results in Figure 9 further show that the non-uniqueness in $P^c - S$ curves is caused by not only the presence of micro-heterogeneities but also the way these heterogeneities are distributed in the domain. HP1 and C4 have the same intensity of heterogeneity (0.25) but they result in different curves and S_{iw} . This is because they contain different distributions of micro-heterogeneity. The R1 pattern has a value of $\omega \sim 0.2$ that is very close to those of both HP1 and C4. Pattern R2 has slightly lower intensity of heterogeneity ($\omega = 0.1$) than all HP1, C4 and R1. Heterogeneities are distributed randomly in R1 and R2 domain. These random distributions seem to have a huge effect on two-phase flow behaviour, distribution of saturation, entrapment of wetting phase in fine sand blocks. They result in different capillary pressure curves and higher S_{iw} values in R1 and R2 domains than HP1 and C4. R5 and R6 domains have very close intensity of randomly distributed heterogeneity in the domain. But, as compared to C4 and HP1, the values of ω in R5 and R6 cases are much less (almost 1/5 of ω in HP1 and C4) and the capillary pressure curves and S_{iw} values are also very different. There is a larger surface area when the heterogeneities are widely distributed so that there will be higher S_{iw} . Although R6 has a lower intensity of heterogeneity compared to HP1, it results in close S_{iw} value to HP1 because of more wetting phase entrapment and also slower flow because of the presence of randomly distributed micro-heterogeneities (pooling effects). Our results also indicate that in random heterogeneity patterns, the domains with close values of ω (e.g., R5 and R6 cases) may result in different capillary pressure curves and S_{iw} value. However, in uniform heterogeneity distribution, the $P^c - S$ curves and S_{iw} value are more or less similar for similar intensity of heterogeneity.

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3.4 Effects of flow directions on effective P^c -S relationships in randomly heterogeneous patterns

We have also looked at the effects of the orientation of a 2D heterogeneous porous domain on P^c -S curves. In general, the work in this area has dealt primarily with samples orientated in a horizontal plane (Ataie-Ashtiani et. al., 2001. 2002; Das et. al., 2004; Manthey et. al., 2005) so as to neglect the effects of gravity on the simulation results. In this section, we analyse the coupled effects of gravity and random micro-heterogeneities on $P^c - S$ curves. Simulations have been carried out on the seven domains R1-R7 and background coarse sand. Each of these samples has been orientated in the North-South (horizontal) plane where gravity effects are the same everywhere as well as on the top-bottom plane (vertically). As discussed before, PCE is then injected from the North (horizontally), top (vertically downward) and bottom (vertically upwards) of the domains.

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The results show a consistent trend in all the patterns R1-R7 (Figure 10). However, a slight discrepancy in our simulations must be pointed out here. In the simulations, the boundary conditions have been altered so that the DNAPL is injected from the bottom but the domains are not turned upside down. This means that

there could be instances where, because of the distribution, there are differences in the levels of trapped water or DNAPL, due partly to the direction of injection rather than solely to gravitational forces. Take for example the case of a hypothetical U-shaped heterogeneity presented in Figure 1. If injection takes place both from above and below, then in the first case there might be a large trapping of DNAPL in the middle of the bend whilst in the second, the heterogeneity may protect the water within the bend from infiltration. This may lead to a significantly different S_{iw} in the domain, especially in a small sample. However such effect does not seem to have had a significant effect on the results for the patterns chosen (R1-R7). Examination of these patterns shows this to be an unlikely scenario in this work. In fact, the main significance of not reversing the pattern is that the distribution of heterogeneity is altered and this does in fact lead to a slight difference in P^c -S curves. However, this confirms again our earlier hypothesis about the effects of distribution on the infiltration process.

Figure 10 presents the derived P^c -S curves for the patterns R1-R7 for injection at horizontal (injection on horizontal plain) and top-bottom planes (injection at both top and bottom of domain). In this figure, R6 pattern is ignored because of the similarity of P^c -S curves in this domain with those in R5 and R7. In patterns R2-R7, the minimum value of S_{iw} occurs when injecting from the bottom and the maximum value of S_{iw} occurs when injecting from the top. However, this trend is not observed in the results for the pattern R1. Instead of the S_{iw} being at a maximum when injecting from the top our simulation gives the minimum value of S_{iw} . The difference between the maximum and minimum S_{iw} is 0.181 compared with a maximum difference of 0.077 in the other domains. We believe this is due to the distribution of heterogeneity in the domain and how it affects flow behaviour. In general, there is a complex interplay of variables which determines the shape of P^c -S curves, hence the amounts of irreducible wetting phase saturation. In the R1 model, the distribution of random heterogeneities affects the fluid front movement, flow path/behavior, saturation distribution and, subsequently the amount of residual saturation. Injection at the top of the R1 model results in pooling of PCE at the top of the random heterogeneities because of higher entry pressure of fine sand blocks, especially toward the left side of the model. When pressure imposed by this DNAPL pool overcomes entry pressure and invades the low permeability fine sand, this adds extra force to pressure forces to displace by-passed/trapped wetting phase in the domain. In case of injection at the bottom, only pressure forces will displace wetting phase since gravity forces act against the displacement direction. Also, heterogeneities are distributed in such a way that upward displacement experiences more low permeability regions than downward displacement. Therefore in the R1 model, contrary to other models, upward displacement results in higher residual saturation compared to downward displacement. This is an effect of how the random heterogeneities are distributed.

The DNAPL saturation plots for various cases are presented in Figures 6(a-b)-7(a-d) for the R2T, R5T, and R6T and Figures 11-12 for the R2B and R2N. The differences in saturation distribution because of the difference in the directions of DNAPL flow are evident in these figures.

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Mitlin et. al. (1998) have carried out a theoretical study of steady-state vertical two-phase flow experiments in a porous material. Their simulation results have indicated that the error in estimating relative permeability and capillary pressure from steady-state vertical flow measurements caused by neglecting gravitational forces is small even for high permeability (10^{-10} m^2) samples. Therefore, they have concluded that if the relative permeability tests are performed on vertical porous domains, gravitational forces can be neglected. This is perhaps plausible since Mitlin et. al. have used a non-wetting phase fluid which is less dense than water and they have the same viscosity values. However, our results for DNAPL flow for the material properties in Table 2 are in contrast to their results. We show that for DNAPL flow, gravity effects are important in determining the P^c -S- K_r curves. Therefore the consideration of whether the gravity effects are included in the simulation or not become important, as this causes non-uniqueness in the capillary pressure relationships.

3.4.1 Haines jumps

When two immiscible phases (DNAPL and water in this case) are brought into contact with a homogeneous porous medium, at equilibrium condition neglecting gravity, the capillary pressure across the interface separating the two fluids is the same everywhere. However, the effective capillary pressure derived by averaging capillary pressure of individual nodes within the domain indicates that this condition is not accomplished and humps appear in the capillary pressure curves. This is because some wetting phase becomes trapped in small pores and around points of contacts between particles in our drainage displacement model. Once hydraulic communication via bulk water path is lost, volumes of disconnected water elements can not adjust quickly to attain equilibrium considering externally applied pressure by both flowing phases. The non-wetting phase pressure slowly increases as drainage progresses until an unstable fluid configuration cause a rapid pressure change. This results in unsteady, impulsive advances of the front of nonwetting phase in the porous medium and this pressure change occurs at almost constant saturation. The sudden motions of unstable interface by small changes in pressure stemmed from unstable fluid configuration have been described as Haines jumps (Haines, W.B., 1930; Miller, E.E., and Miller, R.D., 1956, Melrose, J.C., 1965; Morrow, N.R., 1970 and Mohanty, K.K., 1987). This unstable front movement becomes more complicated in heterogeneous media. This is one of the reasons for humps in P_c -S curves in most of the figures considering either homogeneous or heterogeneous domain. Therefore, due to Haines jumps, the effective capillary pressure does not increase smoothly and monotonously with saturation. As drainage progresses, increasing amounts of water are isolated until irreducible water saturation is reached. At this stage only continuous non-wetting phase and hydraulically discontinuous water are retained. Hence, capillary pressure measured at the domain boundary bears no relationship to the capillary pressure of DNAPL-water interface within individual node across the domain.

However, when capillary pressure is measured experimentally in a porous medium, the discontinuities in pressure are generally too small to observe, and experimental capillary pressure curves measured using inflow and outflow boundary pressure of wetting and non-wetting phase pressure data appear to be smooth and continuous.

3.5 Effects of viscosity ratio on effective P^c -S relationships in randomly heterogeneous patterns

We analyse the effects of altering the viscosity ratio on the P^c -S curves in this section. For the purpose of this paper, the viscosity ratio is defined as the ratio of the viscosity of DNAPL to the viscosity of water, i.e., $\nu = \frac{\mu_{nw}}{\mu_w}$. The domain used for this purpose is the pattern R1 and it has been orientated in the North-South plane (horizontal), thus negating any gravity effects. We have carried out a number of simulations with different values of ν . The original values of μ_{nw} and μ_w are 0.9×10^{-3} and $1 \times 10^{-3} \text{ kgm}^{-1}$, respectively (Table 2) and so the value of ν in our previous simulations is approximately 0.9. For the purpose of this section, ν has been varied between 10 and 0.0001. The other parameters used in each simulation remain identical apart from the variation in the viscosity of the DNAPL.

A typical result from this set of simulations is shown in Figure 13. The value of ν in this case is 0.0001, the lowest value of ν used. As can be seen, this has negligible effect on the P^c -S curves. The DNAPL saturation plots for R1N with two viscosity ratios have also been compared and found to be very similar as can be seen in Figure 14. This implies that capillary forces dominate the viscous forces in this case (zero gravity effect) and is the primary force, which determine the P^c -S curves and saturation distribution of DNAPL. The viscosity ratio does have an effect on the simulations, shown by the fact that the number of time steps taken to reach equilibrium varies between simulations depending on the viscosity ratio. However, this effect does not alter the steady state P^c -S curves. Rose (1960) has shown that the effect of the viscosity ratio on steady state relative permeability to be a second order importance. Our finding backs up this conclusion and we find that when determining DNAPL flow properties viscosity ratio effects are not of significant importance as compared to other more significant factors such as capillary pressure, fluid density, distribution of heterogeneities, pore size distribution, entry pressure of the medium, etc. Therefore, the viscosity ratio may not necessarily cause non-uniqueness in steady state P^c -S curves for DNAPL, if all other factors remain the same.

3.6 Analysis of effective K_r -S curves in randomly heterogeneous patterns

As mentioned before, K_r -S curves quantify the relationships between the relative permeability of the sample to the wetting and non-wetting phases, respectively, at different levels of saturation. They are important in the study of two-phase flow because they effectively determine how ‘easy’ or ‘difficult’ it is for a NAPL to flow through a sample at a given saturation values. Figure 15 shows typical K_r -S curves obtained when injecting from the north, top and bottom of the R3 domain. There are several observations that can be made from these curves. It is clear that gravity has a noticeable effect on the shape of the relative permeability curves. When injecting PCE from the bottom of the sample, the relative permeability of the domain to DNAPL is much less, implying that flow of DNAPL is not so easy. Although relative permeability-saturation changes from 1 to 0, the difference is that, when injecting from the bottom, K_{rnw} decreases faster than injection from the top or horizontally. In this case, The K_r -S curves also show that the relative permeability of the wetting phase drops to small value, well before the S_{iw} is reached and well

before the relative permeability of the non-wetting phase approaches unity. This is particularly clear when injecting from the bottom where it can be seen that the sample becomes almost impervious to the wetting phase at a wetting phase saturation of just under 0.5, whilst becoming impervious to the DNAPL phase at a DNAPL saturation of just under 0.05. They also show that when injecting from the bottom, the sample remains permeable to the DNAPL until a much higher capillary pressure is reached. The inserts in the figures show the shape of the K_{rw} at low wetting phase saturation and it can be seen that the curves follow the expected shape.

4. Conclusions

The present work has focused on characterizing the effects of the interplay between the distribution and intensity of micro-heterogeneities on effective two-phase flow properties of porous media. The effects of gravity on the simulations have been also considered. Our work has concentrated on heterogeneity patterns where fine sand blocks are present in coarse sand. However the patterns have different distributions and intensities of heterogeneities.

It has been found that the intensity of heterogeneity has the greatest effect on the effective two-phase flow properties of porous media. In general, the greater is the intensity of heterogeneity of a sample, the higher is the irreducible water saturation (S_{iw}). However, the distribution of heterogeneity is also found to have influence on S_{iw} and the effective P^c - S - K_r relationships defining the pattern of infiltration.

The effect of gravity has been studied. The general trend observed is that when injecting from the top the S_{iw} tend to be around 15 - 20% higher than when the flow is in a horizontal plane. When injecting from the bottom, the converse is true and the S_{iw} tends to be 15 - 20% less. This effect is obviously a result of gravitational forces.

Our results clearly show that the presence of micro-heterogeneities affect the steady-state P^c - S curves. These are the main input for upscaling flow behavior from core scale to the field scale, either to design oil recovery methods or soil remediation techniques. Therefore, special attention must be taken not to ignore these micro-scale heterogeneities; otherwise the results of field scale simulation depending on the nature, intensity, and distribution of heterogeneities may not be correctly estimated. For example, this may result in erroneous history-matching in oil fields for field development management or evaluation of the efficiency of soil remediation techniques. Therefore, tests must be carried out, e.g. CT scanning rocks or other radioactivity capturing and resistivity logging tools such as gamma ray logging, to determine the nature and distribution of the heterogeneity to display saturation distribution during and at the end of fluid flow cycles.

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729 gravity (Bottom (B))

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Table 1. Intensity of Heterogeneity¹ (ω) of patterns R1 – R7 in Figure 1

Pattern	R1	R2	R3	R4	R5	R6	R7
$\omega(-)$	0.21	0.10	0.104	0.107	0.056	0.059	0.057

¹ the ratio of the total area of micro-heterogeneity to the total sample area

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737

Table 2. Properties of wetting and non-wetting fluids and, coarse and fine sands

Property	Coarse Sand	Fine Sand	Water	PCE (DNAPL)
Permeability, k (m^2)	5×10^{-9}	5×10^{-12}	-	-
Porosity, ϕ (-)	0.40	0.40	-	-
Displacement Pressure, P^d (Nm^{-2})	370	1325	-	-
Pore size distribution index, λ (-)	3.86	2.49	-	-
Irreducible Water Saturation, S_{iw} (-)	0.078	0.098	-	-
Density, ρ (kgm^{-3})	2630	2650	1000	1630
Viscosity, μ (kgm^{-1})	-	-	1×10^{-3}	0.9×10^{-3}
Surface Tension, σ (Nm^{-1})	-	-	0.072	0.035

Figure 1. Random Heterogeneity Patterns (R1-R7) adopted in this work for numerical simulations of two-phase flow in porous media. Each heterogeneous domain is $0.12 \text{ m} \times 0.12 \text{ m}$ is size. The values for the intensity of heterogeneity (ω) in each pattern are shown in Table 1. The U-shaped heterogeneity is used to explain the coupled effects of flow directions and the orientation of heterogeneities on two-phase flow.

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Figure 5. Comparison of P^c -S curves in various random heterogeneity patters (R1-R7) on top-bottom plane with injection of PCE from bottom (B) of domain (vertically upward against gravity). The curves form distinct groups according to the intensity of heterogeneity (ω).

Figure 6. PCE saturation distribution in random heterogeneity pattern R2 for PCE injection along gravity (top of domain) at boundary P^c (a) 400 Pa, (b) 11000 Pa

(a) R5T at $P^c=400$ Pa

(b) R5T at $P^c=1100$ Pa

(c) R6T at $P^c=400$ Pa

(d) R6T at $P^c=1100$ Pa

Figure 7. PCE saturation distribution for injection at the top of random heterogeneity pattern R5 (a-b) and R6 (c-d)

HP1

C4

Figure 8. Representative heterogeneous patterns with uniformly distributed fine sand block in coarse sand, HP1 and C4. Patterns HP1 and C4 have equal intensity (0.25) but different distribution of heterogeneity (adopted from Das et. al., 2004)

Figure 9. Comparison of P^c -S curves for uniformly and randomly distributed heterogeneity patterns with PCE injection from North side (N) of porous domains

(a) Random 1 ($\omega \approx 0.2$)

(b) Random 2 ($\omega \approx 0.1$)

(c) Random 3 ($\omega \approx 0.1$)

(d) Random 4 ($\omega \approx 0.1$)

(e) Random 5 ($\omega \approx 0.05$)

(f) Random 7 ($\omega \approx 0.05$)

Figure 10. Comparison of P^c - S curves for various patterns (R1-R5 and R7) with different intensity of heterogeneity (ω) for PCE flow on horizontal plane (N), along gravity (T) and against gravity (B)

Figure 11. PCE saturation distribution in R2 pattern for PCE injection against gravity (bottom of domain) at boundary P^c of (a) 400 Pa, (b) 11000 Pa

Figure 12. PCE saturation distribution in R2 pattern for PCE injection in horizontal plane at boundary P^c (a) 400 Pa, (b) 11000 Pa

Figure 13. Comparison of P^c - S curves for pattern R1 with different viscosity ratio (ν) for PCE-water system

Figure 14. PCE saturation distribution for pattern R1 with PCE injection in the horizontal plane (North) with viscosity ratio of (a) 0.9 and (b) 0.0001

Figure 15. Comparison of K_r - S curves for water as wetting (w) and DNAPL as non-wetting (nw) phase in patterns R3 where PCE is injected in the horizontal plane (North (N)), along gravity (Top (T)) and against gravity (Bottom (B))