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# Pore scale modelling of DNAPL migration in a water-saturated porous medium

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## Abstract

A numerical simulator based on the discrete network model approach has been developed to simulate drainage processes in a water-saturated porous medium. To verify the predictive potential of the approach to simulate the unstable migration of a dense nonaqueous phase liquid (DNAPL) at the pore scale, the numerical model was applied to laboratory experiments conducted on a sand-filled column. The parameters relative to pore body size and pore throat size used in the construction of the equivalent network were derived from discrete grain-size distribution of the real porous medium. The observed water retention curve (WRC) was first simulated by desaturation of the network model. The good agreement of the modelled WRC with the experimental one highlights that the applied approach reproduces the main characteristics of the real pore space. The numerical model was then applied to rate controlled experiments performed on a homogeneous sand-filled column to study the gravity-driven fingering phenomenon of immiscible two-phase flow of water and a DNAPL. The numerical results match within 10% based on the standard deviation with the experiments. They correctly reproduce the effect of several system parameters, such as flow mode (upward flow and

26 downward flow) and the flow rate, on the stability of the water/DNAPL front in a saturated  
27 porous medium.

28 **Keywords:** network model; porous medium; WRC; DNAPL; immiscible displacement;  
29 fingering

30

## 31 **1. Introduction**

32 Characterization and quantification of the migration of immiscible liquids in groundwater  
33 are topics that have received considerable attention in recent years (Alexandra et al.2012; Wu  
34 et al.2017). Several studies of two-phase immiscible flows have been motivated by the need to  
35 treat spills and leaks of so-called dense nonaqueousphase liquid (DNAPL), which can severely  
36 impact the quality of subsurface water supplies (Kueper and Frind 1989; Birovljevic et al. 1991;  
37 Fayers et al. 1996; Bettahar et al. 1999). When infiltrating throughthe saturated zone, migration  
38 of DNAPLcan result in highly fingered fluid distributions. The occurrence of fingering is  
39 caused by flow instabilities due to differences in viscosity and density between DNAPL and  
40 water (Khataniar and Peters 1992; Riaz and Tchelepia 2006; Essaid et al. 2015; Cheng et al.  
41 2016). These fingers propagate rapidly, causing early breakthrough relative to stable  
42 displacement (Brailovskyet al. 2006).

43 Numerical modelling of DNAPL displacement in water-saturated porous media using  
44 continuum models has been performed by many researchers (Nayagum et al. 2004;  
45 Aggelopoulos and Tsakiroglou 2009, Zhang et al. 2011; Erning 2012; Kokkinaki et al. 2013;  
46 Sleep et al. 2015, Schneider et al. 2015). However, the fingering process in a homogenous  
47 porous medium cannot be simulated with these models because they do not explicitly consider  
48 the instability behaviour of the DNAPL-water interface.

49 To overcome this shortcoming, microscopic approaches such as Stokes' solution for flow  
50 in single pores with a specified geometry or lattice Boltzmann methods(LBM) have been

51 investigated to reproduce unstable displacement experiments. Kiriakidis et al. (1991) developed  
52 a 2D algorithm involving Monte Carlo decision-making, random walks, and principles of  
53 percolation theory. This algorithm successfully predicted the three distinct behaviours of  
54 immiscible displacement in porous media, such as viscous fingering, stable displacement, and  
55 invasion percolation, but the model did not include the gravity term, and gravity fingering was  
56 not considered. Ewing and Berkowitz (1998) developed a generalized growth model based on  
57 invasion percolation to simulate immiscible displacement in saturated porous media. Even  
58 capillary, viscous and gravity forces were incorporated in their 3D algorithm, where viscous  
59 forces were expressed stochastically rather than explicitly. Glass et al. (2001) developed a  
60 macro-modified invasion percolation (MMIP) model, including also the effects of all 3 forces  
61 within the invading phase in a macro-heterogeneous porous medium. The simulated evolving  
62 invasion structure during the gravity-destabilized condition matched the experimental  
63 distributions quite well. Dong et al. (2011) proposed a LBM model taking into account both  
64 wettability and gravity to successfully reproduce flow experiments in Hele Shaw cells. Recently,  
65 Liu et al. (2013; 2014; 2015) have developed a Lattice Boltzmann model that was able to  
66 simulate the main flow regimes (capillary fingering, viscous fingering and stable displacement).  
67 The model was applied to simulate drainage micromodel experiments of the liquid CO<sub>2</sub>  
68 displacement of water. The authors obtained a good agreement with the experimental  
69 measurements and showed that increasing  $M$  (viscosity ratio of non-wetting fluid to wetting  
70 fluid) or decreasing the media heterogeneity can enhance the stability of the displacement  
71 process.

72 While the lattice Boltzmann method gives a more rigorous description of different  
73 physical processes, it is often limited to relatively simple physical situations or to systems  
74 encompassing only a few pores. For this reason, pore-scale networks are often used to describe

75 the flow rate of fluids at the pore scale (Koplick and Lasseter 1985; Aker et al. 1998; Helge et  
76 al. 1999; Jia et al. 1999; Laroche and Visika 2005).

77 In pore-scale network modelling, local capillary equilibrium via the Young-Laplace  
78 equation is generally used to determine multiphase fluid distribution. A set of rules that  
79 describes the appropriate physical processes and arrangements of fluid within each pore is  
80 developed and then combined to describe the displacement of both fluids in the system.  
81 Macroscopic behaviours of the displacement can then be seen in network models, even though  
82 they are not coded explicitly.

83 Many network models have been developed to study a wide range of displacement  
84 processes, including drainage and imbibition (Nordhaug et al. 2003; Singh and Mohenty 2003;  
85 Ferer et al. 2007; Niaser et al. 2009; Tørå et al. 2012; Sheng and Thompson 2013; Bultreys et  
86 al. 2015; Kallel et al. 2017). Regular lattice structures are generally used, with pore bodies  
87 corresponding to the vertices of the lattice and pore throats connecting the pore bodies. The  
88 most important aspects of network models are the dimension and coordination (Gao et al. 2012;  
89 Jivkovet al. 2013). For example, trapping is a dominant process in 2D, less important in 3D,  
90 and virtually non-existent in 4D (3D+time) and when an above multi-directional pore-network  
91 is used (Raouf and Hassanizadeh 2010). Likewise, networks with high coordination behave  
92 quite differently than networks with low coordination. In most early network models, pore  
93 space parameters were chosen without reference to a realistic porous medium. Network is  
94 equivalent only in a statistical sense to the modelled system. Indeed, they were used to explore  
95 network concepts as much as to explore porous media. More modern network models were  
96 often built directly from micro-CT scans of the rock, and so they had not only the same pore  
97 size distribution but also the same exact structure as the rock they are simulating (Al-Raoush  
98 and Willson 2005; Prodanovic et al. 2007; Dong and Blunt 2009; Raouf et al. 2010; Bultreys et  
99 al. 2015). In these cases, the ability to reproduce physical measurements is excellent. However,

100 if the exact rock type has to be used for the network construction, the application of predictive  
101 pore-scale modelling will be severely limited due to the complexity and cost of the methods.  
102 More details about the methods for obtaining pore space information, constructing pore  
103 networks and the application of pore network models in porous media can be found elsewhere  
104 (see Joekar-Niasar and Hassanizadeh(2012) and Xionget al. (2016)).

105 By controlling parameters such as pore-size disorder, viscosity, and flow rate, previously  
106 developed dynamic pore network models were used successfully to reproduce viscous fingering  
107 phenomena exhibited in laboratory studies. Lenormand et al. (1988) developed a dynamic  
108 drainage pore network model and used it to simulate corresponding micromodel experiments.  
109 Their model reproduces capillary fingering, viscous fingering and frontal displacement regimes  
110 for different capillary number and mobility ratio combinations successfully. The largest  
111 network that they used in their simulations consisted of  $100 \times 100$  nodal junctions and they  
112 presented a relatively limited set of simulation results. Aker et al. (1998) developed a two-  
113 dimensional network simulator and used it to characterize the different flow regimes observed  
114 in drainage. They introduced a new method that allowed the simultaneous flow of two liquids  
115 into one pore and demonstrated that viscous fingering could be reproduced. However, the  
116 approach was highly CPU intensive and a  $60 \times 80$  network was the largest lattice that could be  
117 considered. In addition, they only presented results before a breakthrough of the injected  
118 phase. Singh and Mohanty (2003) developed a dynamic network model providing 3D results  
119 consistent with the 2D results of Lenormand et al.(1988). Their simulator could reproduce the  
120 displacement front structure and dynamics as function of capillary number and viscosity ratio.  
121 Whilst their simulator could produce various displacement regimes, including viscous  
122 fingering, the largest system used in their study was a  $30 \times 8 \times 8$  network. Tørå et al. (2012)  
123 extended the model developed by Aker et al. (1998) by incorporating the dynamics of the  
124 wetting layers using an approach similar to Singh and Mohanty (2003). They used this model

125 to study saturation profiles during imbibition and the resistivity index at different capillary  
126 numbers. Their numerical results were in good agreement with experimental data from  
127 sandpacks. The simulations were carried out on one realization of model volume of  
128  $(4.5 \times 1.5 \times 1.5) \text{ mm}^3$  consisting of 767 nodes and 1750 tubes.

129 Most of the previous pore network modelling studies described above were performed in  
130 small networks and were followed by a successful benchmarking exercise of the numerical  
131 simulations against micromodel experiments. However, the modelling approach is rarely used  
132 to simulate such mechanisms at the laboratory core scale. Additionally, very few studies have  
133 presented saturation maps after breakthrough under unstable conditions. An integrated effort is  
134 needed to connect network models and laboratory tests at the core scale more closely.

135 The main objective of the present work is to test, at the scale of a few centimetres, the  
136 capacity of the pore scale network model developed by Nsir and Schäfer (2010) to capture the  
137 gravity-driven fingering observed in our own water-DNAPL drainage system. The model is  
138 first applied to predict the water retention curve of the model sand used in the experiment. The  
139 comparison of the experimental curve and the one modelled will show whether the used  
140 network model reproduces the main characteristics of the real pore space. In a second step, the  
141 numerical model is then used to simulate the displacement of water by DNAPL during  
142 controlled drainage experiments performed in a 68-cm long glass column filled with the  
143 homogenous model sand. The numerical results are compared to the observed physical  
144 quantities, such as the distribution of arrival times of the DNAPL/water front at a control  
145 section, the DNAPL pressure at the inlet as a function of time, and the DNAPL saturation profile  
146 at the end of the displacement experiment.

147

## 148 **2. Laboratory experiments on DNAPL migration**

149 Laboratory experiments were conducted on a 68-cm long glass column with an internal  
150 diameter of 10 cm. The used porous medium is a medium-sized quartz sand with a mean grain-  
151 size diameter of 0.4 mm, a uniformity coefficient of 2.1 and a low fraction of organic content  
152 ( $f_{oc} = 0.09\%$ , based on NFT 31–109) (Bohyet al. 2006). The hydraulic properties of the quartz  
153 sand, such as its moisture characteristics, were quantified in former studies by Cotel(2008).  
154 Trichloroethylene (TCE) was chosen as the DNAPL for the experiments because it is among  
155 the most frequently detected contaminants in subsurface environments. At 20 °C, TCE has a  
156 density of 1.463 g/cm<sup>3</sup>, a viscosity of 0.0056 g/s/cm, and a solubility in water of 1300 mg/L.

157 The experimental setup was designed to produce a controlled and well-defined injection  
158 of TCE in an initially water-saturated homogeneous porous medium. The outlet section of the  
159 column was kept at a constant water pressure. A highaccuracy peristaltic pump with flow rates  
160 up to 200 mL/min was used to inject the displacing fluid in the sand-filled column. Pressures  
161 at the inlet and outlet were measured using a pressure transducer (Cerabar T-PMC131). Several  
162 system parameters (e.g., flow rate, flow mode (upward flow, downward flow) were varied in  
163 the experimental programme.

164 The experimental approach is original, as it is based on the use of optical fibres to locally  
165 quantify the arrival times of the DNAPL/water front at different points within a control section  
166 of the porous medium. Local DNAPL saturations at different depths of the sand column were  
167 also quantified by in situ soil sampling. Because of the emplacement of fibres, the sampling  
168 was achieved only above the sensor control section of the column (between  $z=0$  and  $z= -34$   
169 cm). At each sampling depth, one sample was taken on the axis of the column and two others  
170 on opposite points of the edges of the column section. More details about the experimental setup  
171 and sampling method are given in Nsiret al. (2012).

172

### 173 **3. Numerical model**

### 174 **3.1. Geometry of the network**

175 A 3D network model based on spherical porebodies and cylindrical porethroats was  
176 developed to simulate drainage processes in a saturated porous medium. The pore space is  
177 represented by a cubic lattice with porebodies corresponding to the vertices of the lattice and  
178 porethroats connecting the porebodies. The connectivity or coordination number in the network  
179 (the number of throats connected to a given pore in the network) is set equal to 6. The pore  
180 body and pore throat radii were drawn at random using a probability frequency distribution  
181 given by a special case of the Weibull distribution (Weibull, 1951). We used this type of  
182 distribution because it is easy to implement, and its model parameters may adequately represent  
183 a given pore size distribution. Compared to other models, such as normal, lognormal or bimodal  
184 distribution, the probability of finding smaller pore bodies in the network is higher. In the  
185 modelling approach, specified values of the minimum, mean and maximum radii ( $r_{\min}$ ,  $r_{\text{mean}}$ ,  
186 and  $r_{\max}$ , respectively) are needed to achieve the pore size distribution. The probability  
187 distribution used is as follows:

$$188 \quad g(x) = \begin{cases} \frac{x}{x_2^2} \exp\left(-\frac{x^2}{x_2^2}\right) & \text{for } x \leq x_3 \\ 0 & \text{for } x > x_3 \end{cases} \quad (1)$$

189 where the radius is  $r$ ,  $x = r - r_{\min}$ ,  $x_2 = r_{\text{mean}} - r_{\min}$ , and  $x_3 = r_{\max} - r_{\min}$ . Both porebody and  
190 porethroat radius distributions are of this type, although with different parameters:  $r_{\min}$ ,  $r_{\text{mean}}$   
191 and  $r_{\max}$ . For a throat connecting pores  $i$  and  $j$ , the throat length is defined as  $L_{ij} = L - (r_{\text{pore},i} +$   
192  $r_{\text{pore},j})$ , where  $L$  is the distance between the centres of two adjacent pore bodies.  $L$  is considered  
193 constant in the chosen network. The values of  $r_{\min}$ ,  $r_{\text{mean}}$ , and  $r_{\max}$  specified for both pore body  
194 and pore throat and thus for the node spacing  $L$  are hereafter considered as the geometric  
195 parameters of the network model.

### 196 **3.2. Governing flow equations**

197 When solving the transient flow field, the network model takes into account capillary  
 198 variations in space, the local viscous pressure field, and gravity. The location of fluid-fluid  
 199 interfaces is restricted to the connection between the pore body and pore throat. The Washburn  
 200 equation (Washburn, 1921) is used to quantify the flow rate in a pore throat, which is connected  
 201 to a pore body separated by a water-DNAPL interface. The main assumptions are as follows:  
 202 (i) fluids used are Newtonian, incompressible, and immiscible; (ii) flow is laminar; and (iii)  
 203 local capillary pressure in a pore body is negligible, resulting in only one mean pressure within  
 204 a pore body independent of the phase saturation of that pore body. The mass balance equation  
 205 for each fluid phase  $\alpha$ , wetting (water,  $\alpha = w$ ) and non-wetting (DNAPL or air,  $\alpha = nw$ ), in a pore  
 206 body  $i$  is as follows:

$$207 \quad V_i \frac{\partial S_i^\alpha}{\partial t} + \sum_j Q_{ij}^\alpha = Q_i^{\alpha, \text{inlet}} \quad (2)$$

208 where  $V_i [L^3]$  represents the volume of pore body  $i$ ,  $S_i^\alpha [-]$  represents local saturation (percent of  
 209  $V_i$  filled with fluid  $\alpha$ ),  $Q_{ij}^\alpha [L^3 T^{-1}]$  is the phase flow rate through a pore throat connecting pore  
 210 body  $i$  and pore body  $j$ .  $Q_i^{\alpha, \text{inlet}} [L^3 T^{-1}]$  is the external phase flow rate into pore  $i$ , which is zero,  
 211 unless the pore is located at the flow boundary. The sum runs over all neighbouring pores of  
 212 pore body  $i$ .

213 Summation of Eq. (2) over the two phases gives the following equation for each pore  
 214 body  $i$ :

$$215 \quad \sum_j (Q_{ij}^w + Q_{ij}^{nw}) = \sum_j Q_{ij} = Q_i^{nw, \text{inlet}} \quad (3)$$

216 The expression used for the phase flow rate  $Q_{ij}$ , depends on the fluid present in the pore  
 217 throat connecting pores  $i$  and  $j$ , and on the presence of a meniscus at the boundaries between a  
 218 pore body and a pore throat.

219 If no meniscus is present at the pore throat entry, the flow rate  $Q_{ij}$  through the pore throat  
 220 is given by Poiseuille's law, as follows:

221 
$$Q_{ij} = \frac{\pi r_{ij}^4}{8 L_{ij} \mu_\alpha} (P_i - P_j - (z_i - z_j) \rho_\alpha g) \quad (4)$$

222 where  $\mu_\alpha$  [ML<sup>-1</sup>T<sup>-1</sup>] and  $\rho_\alpha$  [ML<sup>-3</sup>] are the dynamic viscosity and the density of the phase fluid  
 223 present in the pore throat, respectively;  $r_{ij}$  and  $L_{ij}$  are the radius and the length of the pore throat,  
 224 respectively;  $P_i$  and  $P_j$  are the fluid pressures in pores  $i$  and  $j$ ; and  $z_i$  and  $z_j$  are the height of the  
 225 vertices at the centre of pore bodies  $i$  and  $j$ . The first term of the right-hand side of Eq. (4),  
 226  $\pi r_{ij}^4 / (8 L_{ij} \mu_\alpha)$ , represents the flow conductance in the pore throat.

227 If a meniscus is present at the pore throat entry, the flow rate  $Q_{ij}$  through the pore throat  
 228 is expressed by the Washburn equation (Washburn, 1921), as follows:

229 
$$Q_{ij} = H \left\langle P_i - P_j - (z_i - z_j) \bar{\rho} g - P_c(r_{ij}) \right\rangle \frac{\pi r_{ij}^4}{8 L_{ij} \mu} (P_i - P_j - (z_i - z_j) \bar{\rho} g) \quad (5)$$

230 where  $P_c(r_{ij})$  [ML<sup>-1</sup>T<sup>-2</sup>] denotes the threshold capillary pressure that must be exceeded for the  
 231 DNAPL to enter the pore throat connecting pore bodies  $i$  and  $j$ . This threshold pressure is given  
 232 by the Young-Laplace equation, as follows:

233 
$$P_c(r_{ij}) = \frac{2\sigma \cos\theta}{r_{ij}} \quad (6)$$

234 where  $\theta$  is the contact angle (assumed to be zero in this work).  $H \langle X \rangle$  is a Heaviside function  
 235 that stands for the displacement condition of the interface in a pore throat. The throat can be  
 236 blocked by capillarity if the capillary entry pressure ( $P_c(r_{ij})$ ) has not yet been reached. The  
 237 Heaviside function is defined as follows:

238 
$$H \langle X \rangle = \begin{cases} 1 & \text{for } X > 0 \\ 0 & \text{for } X \leq 0 \end{cases} \quad (7)$$

239 If a meniscus is present at the boundary between a pore body and a pore throat, the identity  
 240 of the fluid flowing through the pore throat (DNAPL or water, or capillary-blocked) can only  
 241 be determined after computation of the pressure field in the network. Therefore, an effective  
 242 viscosity and an effective density are used for the displacing fluid in Eq. (5) and are defined by

243 the following:

$$\begin{aligned} \bar{\mu} &= \mu_{nw} S_{ij}^{nw} + (1 - S_{ij}^{nw}) \mu_w \\ \bar{\rho} &= \rho_{nw} S_{ij}^{nw} + (1 - S_{ij}^{nw}) \rho_w \\ S_{ij}^{nw} &= (S_i^{nw} + S_j^{nw}) / 2 \end{aligned} \quad (8)$$

245 where  $S_{ij}^{nw}$  [-] is the average phase saturation between pore bodies  $i$  and  $j$ . As the average phase  
246 saturation evolves as a function of time, it indirectly accounts for the interface movement inside  
247 a pore throat.

248 Introducing the appropriate expression of flow rate  $Q_{ij}$  in Eq. (3) leads to a set of algebraic  
249 equations with the pore body pressures as unknowns. This set of equations is written in matrix  
250 form, as follows:

$$251 \quad \mathbf{A} \times \mathbf{B} = \mathbf{P} \quad (9)$$

252 where  $\mathbf{A}$  is the conductance matrix with elements that depend on the connections between  
253 different throats,  $\mathbf{P}$  denotes the pressure vector, and the  $\mathbf{B}$  vector contains the pressure and the  
254 flow rate at the boundaries, the buoyancy terms, and the capillary pressure if a meniscus is  
255 present in the pore throat.

### 256 3.3. Numerical solution procedure

257 The governing equations presented in the previous section are solved for fluid pressure  $P$   
258 and wetting fluid saturation  $S_w$  using an IMPES (IMplicit Pressure-Explicit Saturation)  
259 modelling approach. The flow chart describing the different steps of the pore-scale network  
260 model is shown in Figure 1.

261 Equation (7) is solved implicitly for  $P$ , assuming that  $S_w$  and throat conductances  $\pi r_{ij}^4 / (8$   
262  $L_{ij} \mu_\alpha)$  are known from the previous time step. The calculated pressure field is then used to  
263 quantify fluid flow rates through the pore throats. The ratio of free volume in a pore body  $i$  (not  
264 invaded by displacing fluid) to the sum of flow rates in all pore throats connected to pore body  
265  $i$  and containing a moving interface gives the filling time needed for that pore body  $i$  to be

266 invaded. The minimum value of the calculated filling times (hereafter called  $T_{\min}$ ) is then used  
267 to update the local saturation in each pore body as follows:

$$268 \quad S_i^{t+1} = S_i^t + \frac{Q_i \times T_{\min}}{V_i} \quad (10)$$

269 where  $S_i^t$  [-] and  $S_i^{t+1}$  [-] denote the saturation at the previous time (t) and calculated at the new  
270 time (t+1), respectively; and  $Q_i$  [ $L^3T^{-1}$ ] represents the sum of flow rates in all pore throats  
271 connected to pore body i and containing a moving interface.

272 During one time step, only one pore body reaches the full non-wetting phase saturation.  
273 This pore body generates additional interfaces at all connecting pore throats that are filled with  
274 the wetting fluid. Newly created interfaces are tested for stability, and appropriate expressions  
275 for flow rate  $Q_{ij}$  are then identified. This modifies the conductance matrix at each time step.  
276 The field pressure is computed, and the procedure for updating the saturation in pore bodies is  
277 repeated. This provides a transient response for the pressure and saturation fields.

278 To solve the set of equations (9), a Preconditioned Conjugate Gradient Method (PCGM)  
279 algorithm is used. The simulations are conducted on a Linux-computer with 3956 MB RAM  
280 and a Celeron processor running at 1998 MHz. One iteration on the chosen lattice network  
281 ( $20 \times 20 \times 200$ ) took approximately 90 seconds and required a memory space of approximately 4  
282 MB. The corresponding Central Processing Unit (CPU) time was approximately twenty days  
283 for the chosen lattice size. As the Fortran code was not parallelised and the free memory offered  
284 by the used computer was limited, simulations could not be launched when a lattice size larger  
285 than  $20 \times 20 \times 200$  was used.

286 The time-stepping process is repeated until a stopping condition is reached. This  
287 condition allows the choosing of the stage at which the simulation should be achieved: after  
288 attaining a fixed saturation value at a given cross-section, after reaching a breakthrough of the

289 invading fluid phase, after having performed a defined number of steps, or after the injecting  
290 of a predefined total volume of the invading fluid phase.

### 291 **3.4. Model parameters and numerical implementation**

292 To obtain a reliable simulation of laboratory experiments, the characteristic parameters  
293 of size distributions for both pore bodies and pore throats ( $r_{\min}$ ,  $r_{\text{mean}}$  and  $r_{\max}$ ) were computed  
294 based on the grain-size distribution (GSD) of the real porous medium. The modelling concept is  
295 based on the geometric properties of packing spheres (Dodds, 1980), which are combined with  
296 a probability approach proposed by Rouault and Assouline (1998). The input data for sphere  
297 packing are in the form of a discrete particle-size distribution derived from the (GSD) of the  
298 sand used in the experiment (Nsir and Schäfer, 2010). The obtained results consist of one pore  
299 body-sized distribution curve and four pore throat-sized distribution curves. Assuming that the  
300 generated pore body sizes and pore throat sizes are normally distributed, which is generally  
301 assumed for a large sampling number, assessment of the mean value and the standard deviation  
302 (sd) of corresponding curves allows the computation of various ranges of pore body radii and  
303 pore throat radii in terms of  $r_{\min}$ ,  $r_{\text{mean}}$  and  $r_{\max}$ . More details of the applied method are given in  
304 the work of Nsir and Schäfer (2010).

305 The geometric parameters for the pore body radii and pore throat radii considered in the  
306 numerical simulations are shown in Table 1. The grid length should be slightly two times greater  
307 than the maximum radius of pores ( $r_{\max}=0.054$  mm). For this reason, the distance between the  
308 centres of two adjacent pore bodies ( $L$ ) was fixed to 0.14 mm. The chosen values gave the best  
309 fit for the measured macroscopic properties of the medium sand used in the experiment. The  
310 equivalent intrinsic permeability and porosity are  $8.6 \times 10^{-11}$  m<sup>2</sup> and 47%, versus the measured  
311 values of  $9 \times 10^{-11}$  m<sup>2</sup> and 40%, respectively (Nsir and Schäfer, 2010). By fixing the maximum  
312 radii ( $r_{\max}$ ) and the minimum radii ( $r_{\min}$ ) of the size distribution as one time, two times and three

313 times the standard deviation (sd) away the mean radius ( $r_{\text{mean}}$ ), three size distributions with  
314 different shapes were considered for a given lattice size (see Table 1).

315 To simulate the entire laboratory column (68 cm long, 10 cm internal diameter), an  
316 equivalent network with  $714 \times 714 \times 4858$  nodes in the [X,Y,Z] directions, corresponding to  
317 2,476,588,968 pore elements is required. Simulations with such lattice size are highly CPU  
318 intensive and RAM consuming and were, consequently, not manageable by our computational  
319 resources. To face this difficulty, we decided to reduce the size of the network in order to match  
320 the maximum height and width of the column.

321 An equivalent network of  $7 \times 7 \times 4858$  pore bodies was initially chosen to simulate drainage  
322 experiments. This spatial discretisation has the advantage of reproducing the total height of the  
323 physical model and, consequently, gravity effects, even when only a small fraction of the  
324 column section is used (1 square millimetre). This might favour the formation of very few  
325 fingers that might occupy most of the cross-section of the model. As they move downward, the  
326 number of pore bodies to invade before reaching the bottom might increase the drainage of the  
327 porous medium. Consequently, an overestimation of the saturation of the invading fluid can be  
328 induced by a coarse pore discretisation in the transverse direction. Using the correct shape of  
329 the cross-section is thus as important as the real column height.

330 To account for a sufficiently large section of the medium that is manageable by our proper  
331 computational resources, additional simulations considering networks with more nodes along  
332 the two directions of the horizontal section ( $20 \times 20$ , rather than  $7 \times 7$ ) but with minimum nodes  
333 in the direction of flow (200, instead of 4858) have also been undertaken. To conserve  
334 representation of the total physical height, the node-spacing was increased (3.4 mm, instead of  
335 0.14 mm). Increasing the node spacing does not impact the macroscopic properties of the  
336 equivalent network as already discussed in the previous work of Nsirand Schäfer (2010).  
337 Furthermore, the corresponding network may be assimilated in this case to a package of

338 cylindrical tubes joining at junctions with volume, which remains the physical representative  
339 of reality.

340 To reproduce the conditions of the performed drainage experiment, the network model  
341 was initially water-saturated and a constant flow rate of  $29.6 \times 10^{-4}$  mL/min of DNAPL was  
342 applied at the inlet section of the model. As the simulated cross-section of the network was  
343 significantly smaller than the real cross-section of the experimental soil column, the simulated  
344 injection rate was reduced to the experimental injection rate by the ratio of the cross-sectional  
345 area of the model to that of the column. At the outlet section of the model, the observed water  
346 head was specified as a constant pressure boundary. The simulation was stopped when the time  
347 needed to inject the DNAPL volume considered in the experiment was reached.

348

## 349 **4. Results and discussion**

### 350 **4.1 Water retention curve modelling**

351 The relationship between the amount of water, typically quantified by the degree of  
352 saturation (S) or volumetric water content ( $\theta$ ), and pore-water potential, typically quantified by  
353 matric potential or capillary pressure ( $P_c$ ), is referred to as the water retention curve(WRC) of  
354 soils. A key attribute of the developed model is its capability to compute fluid pore-scale  
355 saturation as a function of capillary pressure applied to a given lattice, which allows modelling  
356 the WRC of a porous medium. Comparison of the numerically calculated WRC with the  
357 experimentally obtained one allows for verification if the predicted pore space parameters match the  
358 features of WRC observed in a column experiment.

359 The experimental data used for verification were obtained independently in a multistep  
360 outflow experiment (Cotel, 2008). To reproduce the functional relationship between capillary  
361 pressure and water saturation, a displacement simulation was performed using a series of  
362 sequential capillary pressure steps. The water saturation of the porous medium was computed

363 at the end of each displacement step. For each prescribed capillary pressure, the dynamics of  
364 the desaturation of the medium is quantified by the track of the generated interfaces in the pore  
365 network. This rule was applied iteratively until all the air/water interfaces remained at rest under  
366 the given capillary pressure level, and the invasion of water-filled pores by air is stopped. At  
367 this stage, the whole network is under hydrostatic conditions and the corresponding water  
368 saturation can thus be computed. Then, the applied capillary pressure is again increased. The  
369 simulation is stopped when the air/water front reached the outlet face of the network.

370 The simulations were conducted on a network of  $20 \times 20 \times 200$  nodes because it has lower  
371 CPU consumption and presents a larger cross section than the  $7 \times 7 \times 4858$  lattice. Furthermore,  
372 matching a fine vertical discretisation of the column is not absolutely necessary to model the  
373 macroscopic capillary pressure as a function of the water saturation. According to the  
374 experimental data, the first capillary pressure applied to the network boundary was fixed to 100  
375 Pa. The network was then drained stepwise by systematically incrementing the prescribed  
376 capillary pressure by a value of 100 Pa at each displacement step.

377 The simulated WRC for different pore throat size distributions compared to the measured  
378 one are given in Figure 2. The agreement in the sloping of the curves shows saturation changes  
379 in a similar fashion for a given change in pressure. The model seems to be able to adequately  
380 predict the observed capillary pressure-saturation relationship up to a capillary pressure of  
381 approximately  $0.2 \times 10^4$  Pa (equivalent to 0.2 m water head). The model predicts slightly higher  
382 water saturations than those measured in the experiment. However, it clearly deviates from the  
383 experimental data set for high capillary pressures corresponding to water saturations lower than  
384 0.2. The mobilisation of the wetting phase in the simulated network appears significantly higher  
385 than that observed in the experiment in this condition. This may be due to limitations of the  
386 numerical model when a network section of only  $(20 \times 20)$  pores in a horizontal transect is used.  
387 In fact, neglecting the spatial pore size correlations or connectivity may lead to inaccurate

388 predictions of water retention at very low water saturation. In addition, the simulated fluid  
389 displacement is considered frontal. Thus, entrapment of the wetting phase due to snap off is not  
390 considered in the numerical model. This means that water cannot be trapped as the menisci  
391 advance into a pore throat and there is no residual wetting phase left behind.

392         The misfit between the measured WRC and modelled one may also reflect limitations  
393 inherent in the experimental results within this range, where water retention mechanisms other  
394 than capillarity may dominate. Several studies on the WRC of soils led us to believe that the  
395 water retention characteristic is not only closely related to the pore size distribution but also to  
396 the solid pore interfaces (Rossi and Nimmo, 1994; Fayer and Simmons, 1995; Webb, 2000;  
397 Tuller and Or, 2005; Khlosi et al. 2006; Silva et al. 2007). The experimental setup used in the  
398 laboratory experiment does not provide a highly accurate description of the WRC in the very  
399 dry range of the sand. On the other hand, using a wider distribution of the pore throat size gives  
400 a much better match with the measured one (Figure 2). This is clearly due to the generation of  
401 a few additional thinner pore throats when doubling or tripling the width of the distribution.  
402 The penetration of the non-wetting phase into the pore bodies is thus reduced, and more residual  
403 water phase is left behind. The characteristics of the pore throat distribution used in the network,  
404 such as the mean radius and standard deviation, seem to be a key factor to minimise the  
405 difference between experimentally observed WRC and numerically modelled WRC.  
406 Optimizing the distribution parameters in this way is similar to optimising fitting parameters in  
407 commonly adopted WRC models (Brooks and Corey, 1964; Van Genuchten, 1980). However,  
408 the link between modelling parameters and physical properties of the soil is here more explicit.  
409 Despite this improvement, the shape of the network, i.e. the number of pores simulated in the  
410 horizontal transect on the WRC, remains the principal cause of the misfit between the observed  
411 and the modelled WRC.

412 Using a 2D network on a X-Z plane allowed us to study the impact of the network shape  
413 on the numerical results and to compare them to the experimental results, as we are now able  
414 to double or quadruple the number of pores of the network along the cross-section. Such a  
415 choice was not possible with a 3D network, as the computational resources do not permit  
416 running simulations with a large lattice size. Here, we studied the effect of the shape of the  
417 network on the WRC.

418 Simulated water retention curves with networks of  $50 \times 50$ ,  $75 \times 75$ , and  $100 \times 100$  height by  
419 width, but with the same pore throat size (1 sd), are compared with the experimental curve  
420 (Figure 3). Numerical results show now a good agreement with the measured curves, especially  
421 in the region of pendular water ( $S < 0.2$ ). Compared to the 3D simulation (with a small cross  
422 section of  $20 \times 20$ ), the simulated ( $100 \times 100$  based) water retention curve (WRC) fits the  
423 measured WRC quite well at low water saturation but not at high wetting saturation ( $0.2 < S < 1$ ).  
424 This indicates that the larger the horizontal transect is, the higher the trapped water will be. On  
425 the other hand, reducing the vertical discretisation of the pore network will lead to an  
426 underestimation of the quantity of capillary and funicular water. The behaviour of the pore  
427 network is thus shape dependant (function of the number of pores modelled in the cross-  
428 section), as well as size dependant (number of pores modelled along the height). Based on these  
429 results, the discrete network model should have enough nodes in all three directions to obtain  
430 accurate matching of the experimental WRC.

431

## 432 **4.2. Modelling of a laboratory experiment on DNAPL/water drainage**

### 433 **4.2.1. Displacement behaviour of the water/DNAPL front for a low injection rate**

434 The simulations were conducted on a network of  $20 \times 20 \times 200$  nodes in the [X,Y,Z]  
435 directions. This lattice size was preferred to the  $7 \times 7 \times 4858$  lattice size because the modelled

436 cross-section is larger, and as shown in section 4.1, the emergent network behaviour is shape  
437 dependent.

438 The DNAPL arrival times computed at individual pore bodies of a control section as  
439 defined in the experiment setup (Nsiret al. 2012) have been compared to the measured arrival  
440 times for both vertical-upward and vertical-downward flow modes (Figure 4). The  
441 corresponding curves were plotted as a function of DNAPL arrival time, normalized by the  
442 average arrival time for both the experiments and numerical simulations. As shown in Figure 4,  
443 the simulations exhibited behaviours similar to those observed experimentally. In fact, the  
444 dimensionless DNAPL arrival times calculated by the developed model for both flow modes are  
445 in good agreement with those obtained in the experiments. The calculated standard deviations  
446 are also rather close to the measured values. In fact, the standard deviation values were 14 sec  
447 and 75 sec, compared to 9 sec and 70 sec measured in the experiments, for the case of vertical-  
448 upward displacement and vertical-downward displacement, respectively. Thus, the model  
449 is able to adequately reproduce even the observed displacement instabilities caused by the  
450 negative contribution of gravity.

451 In the case of upward displacement, the gravitational component is added to the total  
452 potential driving flow and creates an increase in the global inlet pressure. Consequently, the  
453 fraction of accessible pores increases and a compact pattern of the invading fluid is established,  
454 resulting in a nearly uniform distribution of DNAPL in a porous medium transect. In contrast,  
455 because of the negative effect of the gravity forces, the water/DNAPL interface is no longer  
456 uniformly distributed over the cross-section in the case of downward displacement and many  
457 pore throats become blocked by capillary effects. The number of active invasion paths is  
458 therefore lower in this situation, and many vertically oriented fingers may develop in the pore  
459 network.

#### 460 **4.2.2. DNAPL saturation**

461 The final average DNAPL saturations calculated over the entire network are quite  
462 different for upward and downward displacements, confirming that density contrast plays a  
463 significant role in the migration of a DNAPL in a porous medium, particularly at low DNAPL  
464 injection rates. According to the numerical simulations, we determined an average DNAPL  
465 saturation of approximately 72% in the case of upward displacement but only 41% for  
466 downward displacement. These values are in the same order of magnitude of those obtained in  
467 the experiments as an average DNAPL saturation of 66% and 33% was obtained for upward and  
468 downward displacement, respectively. This difference is mainly due to the stable growth of the  
469 water/DNAPL front during upward displacement, whereas the downward displacement is  
470 characterized by flow instabilities that were obviously reproduced quite well with the developed  
471 model.

472 Figure 5 illustrates a 3D representation of the calculated DNAPL saturation along a 1.5-  
473 cm length of the equivalent network of the medium sand used in the experiment. The numerical  
474 results correspond to an unstable downward displacement. The 3D view of saturation field  
475 clearly illustrates the formation of gravity-enhanced fingering in the simulated porous medium.  
476 In this condition, only a small amount of displaced fluid (water) comes in contact with the  
477 DNAPL and due to the destabilizing effect of gravity, the displacing fluid quickly invades the  
478 larger throats and blocks narrow throats where water remains entrapped. Consequently, the  
479 advancing DNAPL/water front exhibits several fingers that elongate downwards while  
480 advancing through the network.

481 Based on the DNAPL saturation field computed in the network at a given time, a volume-  
482 averaged DNAPL saturation can be computed over a representative volume of the medium. The  
483 resulting macroscopic profiles of volume-averaged DNAPL saturation, as a function of depth  
484 for both upward and downward displacement, were then compared to those obtained in the  
485 experiment (Figure 6). Similar to the experimental observations, the front behaviour of predicted

486 DNAPL saturations appears to be abrupt in the case of upward displacement and presents  
487 remarkable fluctuations in the case of downward displacement. The numerical results confirm  
488 the experimental observations that the water-DNAPL displacement occurs uniformly when  
489 DNAPL is injected from the bottom. However, a heterogeneous cross-section distribution of  
490 DNAPL is obtained when the displacing fluid is injected from the top of the column due to both  
491 gravity effects and an unfavourable viscosity ratio. The numerical results confirm the  
492 experimental findings. However, the calculated DNAPL saturations are higher than the  
493 measured values for both displacement modes (upward and downward), especially when a  
494 narrow pore throat size distribution is used (1 sd). Significant differences appear, particularly  
495 in the region close to the network inlet. For example, at a depth between 10 and 20 cm,  
496 differences in saturation may reach 40% in the case of stable water displacement. Apparently,  
497 the simulated number of active and invading fluid paths seems to be relatively higher than the  
498 experimental values. Discrepancies to the measurement are reduced and a better fit is obtained,  
499 particularly in the case of downward displacement, when a wider pore throat size distribution  
500 is used (case where  $r_{\max}-r_{\text{mean}}= 2$  sd). In this case, much higher capillary pressure in the pore  
501 throats is generated and the possibility to overcome it is significantly reduced. This might  
502 equilibrate interplay between viscous forces, enhancing the full drainage of water, and capillary  
503 forces that are acting to retain displaced fluid. Consequently, the displacement of water by the  
504 DNAPL is now better represented by the model leading to simulated DNAPL saturations that  
505 are closer to the measured ones.

506 As previously mentioned when modelling WRC (see section 4.1), the relatively higher  
507 residual saturations of the invading fluid predicted by the numerical model can also be attributed  
508 to the chosen small cross-section of the model. The simulated cross-section of the equivalent  
509 network is represented by a small number of nodes ( $20 \times 20$ ) and remains small compared to the  
510 reality, which may have limited development of fingering patterns during DNAPL

511 displacement. Using a larger section of network may be more appropriate to model the real  
512 porous medium, but larger networks suffer from prohibitive computation times even with  
513 powerful computers. To show the effect of the cross-section shape on the emergence of DNAPL  
514 displacement behaviour, we have undertaken a series of additional simulations with a short  
515 reduced height (100 nodes) and three sizes of the cross-section,  $6 \times 6$ ,  $12 \times 12$ , and  $24 \times 24$ . Using  
516 a much larger cross-sectional size ( $>24 \times 24$ ) was not manageable with our proper computational  
517 resources. Figure 7 shows the DNAPL saturation field and vertical section-averaged DNAPL  
518 profile calculated after 8 sec of injection for each of the three cross-sections. The larger the  
519 cross-section is, the more advanced the DNAPL fingers are and the less DNAPL saturated the  
520 pores are at the inlet section. This clearly underscores the need to increase the cross-section by  
521 at least 5 times more nodes in each direction to obtain an improved fit of the experimental  
522 results.

#### 523 **4.2.3. DNAPL pressure at the inlet section**

524 Although the DNAPL saturation describes the distribution of displacement fluids in the  
525 porous medium, it is the pressure build-up that significantly determines the displacement of one  
526 fluid by another. Thus, to better understand the interplay between global pressure behaviour  
527 and the displacement condition (stable or unstable), the variation of the inlet DNAPL pressure  
528 as a function of time was predicted by the model and compared to the measurements for the  
529 cases of upward displacement and downward displacement, respectively (Figure 8). According  
530 to the measured data, the boundary pressure (applied to the outlet section of the experimental  
531 system), which was taken into account in the simulation for both upward and downward  
532 displacement, corresponded to a water height of 18 cm and 86 cm, respectively. As observed  
533 in the experiments, the pressure increased with time in the case of vertical upward displacement,  
534 whereas it decreased in the case of downward displacement.

535 In the situation where DNAPL is injected at the bottom of the network, the water/DNAPL  
536 interface is stabilized by the gravity effect and the potentially destabilizing influence of the  
537 viscosity ratio was not significant. The pressure increase is therefore caused by the high density  
538 of the invading fluid, which overrides the decrease in viscous pressure drop during the  
539 displacement of the less viscous DNAPL. Although the time-dependent increase in DNAPL  
540 pressure was well reproduced by the numerical simulations, the calculated pressure increase  
541 seems to be slightly lower than that observed in the experiment. This can be explained by the  
542 chosen arrangement of throat sizes in the network model. Certain narrow throats that may exist  
543 in the real porous medium were not captured in the network and may require a higher  
544 displacement pressure to be invaded by the displacing fluid.

545 In the case of downward DNAPL displacement, the interplay between hydrostatic  
546 pressure and viscous pressure changes leads to an overall decrease of the inlet DNAPL pressure.  
547 The sudden increase of pressure illustrated in Figure 8b corresponds to the breakthrough of the  
548 invading fluid at the outlet section. Here, gravity forces are destabilizing and dominate the  
549 movement, resulting in typical DNAPL fingers in the displaced fluid. The number of active  
550 invasion paths is therefore lower and the flow section of DNAPL is reduced. The contribution  
551 of viscous pressure is also limited and is dominated by the contribution of the higher density of  
552 the invading fluid. The combination of all these effects results in decreased inlet pressure. The  
553 observed decrease of inlet pressure is correctly reproduced by the numerical model, and the rate  
554 at which it decreases seems to be relatively higher than that of the measured one. In addition,  
555 the computed breakthrough time is earlier compared to that observed in the laboratory  
556 experiment. This can be explained by the number of active invasion paths, which may be lower  
557 in the network model than in the real porous medium. The leading fingers as simulated in the  
558 model (see Figure 5) are essentially moving straight, pertaining to a zero tortuosity. The

559 development of such “no wiggle” fingers decreases the number of pore bodies to invade before  
560 reaching the bottom, accelerating the simulated breakthrough.

#### 561 **4.2.4. Influence of the DNAPL injection rate on gravity fingering**

562 The DNAPL injection rate is one of the key factors in drainage processes, and it controls  
563 the balance between buoyancy, viscous forces, and capillary forces. It has been previously  
564 shown (see section 4.2.2) that under a low injection rate, the vertical-downward displacement  
565 of water by DNAPL gives rise to several fingers. The invasion of the saturated porous medium  
566 is only partial, and the resulting distribution of the DNAPL is thus inhomogeneous. Nsiret al.  
567 (2012) showed in previous experimental work that increasing the flow injection rate  
568 significantly stabilizes the displacement and may entirely suppress the fingering process during  
569 immiscible downward displacement. This scenario was also simulated by the developed model  
570 to test its capacity to reproduce such phenomena.

571 Figure 9 shows the comparison of the simulated dimensionless arrival times of the  
572 water/DNAPL front with the measured values. The statistical data of both distributions are  
573 similar. The calculated standard deviation was very close to the measured value (24 sec against  
574 20 sec measured in the experiment) and much lower than that computed at the low injection rate  
575 condition (75 sec). Likewise, the mean arrival time was approximately 489 sec, which is  
576 compared to the observed time of approximately 512 sec. Thus, although gravity fingering is  
577 considered to be a highly dynamic process, the numerical results also show that increasing the  
578 flow rate alters its destabilizing effect and leads consequently to the stabilization of the  
579 water/DNAPL front. At a low injection rate, DNAPL moves through a pathway of larger pore  
580 bodies and pore throats with the lowest capillary entry pressure because the viscous forces are  
581 not significant and do not disrupt the dynamic filling of pore bodies and pore throats. This leads  
582 to a ramified invasion and to a preferential path growth in the saturated porous medium. As the  
583 flow rate increases, viscous forces become more significant, more menisci become mobile, and

584 consequently, even fine pore throats may be as overcome as larger ones because of the  
585 significant pressure gradient. As a result, a compact pattern of the invading fluid occurs during  
586 displacement.

587 Figure 10 illustrates the macroscopic profile of the volume-averaged DNAPL saturation  
588 as a function of depth, calculated when the injection rate of DNAPL was doubled and compared  
589 to the measured one. Here, the DNAPL saturations are higher and their spatial distribution is  
590 clearly more homogenous compared to those calculated under low injection rate. Such  
591 macroscopic behaviour is in good agreement with the experimental observations. However, the  
592 simulation results slightly overestimate the monitored DNAPL saturation. Discrepancies are  
593 significantly reduced when a wider pore throat size distribution is used in the simulation. The  
594 arguments previously detailed in the case of the low injection rate (see section 4.2.2) may also  
595 explain the small misfit between measured and calculated average DNAPL saturations. In  
596 addition, increasing the size of the network cross-section may improve the fit of the numerical  
597 results with the experiment.

598

## 599 **5. Conclusions**

600 The study presents detailed comparisons between numerical simulations of drainage  
601 processes (displacement of water by air, water by DNAPL) and corresponding physical  
602 experiments at the scale of a few tens of centimetres. The goodness of fit between the developed  
603 pore-scale network model and the experiment seems to be a function of the grid characteristics  
604 and modelling domain dimensions. A major feature of the model is its capacity to reasonably  
605 well reproduce the redistribution of the fluid phases in the pore space for a given pressure  
606 change, a modification of the applied displacement condition (stable/unstable) or for a rate  
607 injection increase. In fact, the predicted water retention curve matched well globally with that  
608 obtained in the experiment, especially within the region of higher water saturation ( $0.2 < S < 1$ ).

609 The predicted distribution of arrival times of the water/DNAPL front computed at a given cross-  
610 section are in good agreement with the experimental data obtained for a given displacement  
611 scenario. The slightly higher DNAPL saturations predicted by the numerical model are  
612 essentially attributed to the chosen small cross-section of the equivalent network. In the actual  
613 version of the model, the lattice sizes are limited by the computation time and space memory  
614 and remain too far to adequately well represent the whole cross-section of the real porous  
615 medium. Thus, a network model with enough nodes for the cross-section allows distinct  
616 behaviour of immiscible displacement, such as DNAPL gravity-driving fingering to emerge  
617 sufficiently from the interactions of the various pores and throats that constitute the network  
618 and the various forces taken in account in the model. This may reduce the differences between  
619 experimentally determined saturations and modelled saturations. In this context, further  
620 developments of the numerical code, such as implementation of an iterative and more powerful  
621 solver, should be undertaken to reduce and optimize the CPU time. Complementary studies are  
622 also necessary to test additional features of the pore-space geometry of the porous medium. The  
623 model can also be extended to simulate the imbibition process and promote application of the  
624 model to reproduce the experimentally obtained residual DNAPL saturations.

625

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634

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795 simulation of liquid and gas two phase flow based on digital core technology. *Science*  
796 *China Technological Sciences.* 58, 1375-1384

797 Table 1: Minimum, mean, and maximum radii of pore bodies and pore throats derived from  
 798 the sphere-packing model.

	minimum ( $r_{\min}$ )	0.042		
Pore-body radius [mm]	mean( $r_{\text{mean}}$ )	0.048		
	maximum ( $r_{\max}$ )	0.054		
		$r_{\max}-r_{\text{mean}}=\text{sd}$	$r_{\max}-r_{\text{mean}}=2 \text{ sd}$	$r_{\max}-r_{\text{mean}}=3 \text{ sd}$
Pore-throat radius [mm]	minimum ( $r_{\min}$ )	0.026	0.022	0.018
	mean ( $r_{\text{mean}}$ )	0.030	0.030	0.030
	maximum ( $r_{\max}$ )	0.034	0.038	0.042

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### 801 **Figure captions**

802 Fig. 1: Flow chart of the pore-scale network model. The time-stepping process is repeated until  
 803 a stopping condition is (dynamically) reached, or when equilibrium state is achieved, i.e., all  
 804 the interfaces are blocked.

805 Fig. 2: Experimental and modelled WRC of the quartz sand used in the laboratory experiments.  
 806 Simulations with a 3D lattice network ( $20 \times 20 \times 200$ ) and 3 different pore throat radii  
 807 distributions. The capillary pressure value is expressed as equivalent water head (m H<sub>2</sub>O).

808 Fig. 3: Experimental and modelled WRC of the quartz sand used in the laboratory experiments.  
 809 Simulations with a 2D lattice network with 3 different sizes ( $50 \times 50$ ,  $75 \times 75$ , and  $100 \times 100$ ) and  
 810 a given pore-throat radii distribution ( $r_{\max}-r_{\text{mean}}=1\text{sd}$ ). The capillary pressure value is expressed  
 811 as equivalent water head (m H<sub>2</sub>O).

812 Fig. 4: Measured and simulated dimensionless arrival times of the water/DNAPL front using a  
 813 3D lattice network ( $7 \times 7 \times 4858$ ) in both vertical-upward flow mode and downward flow mode:  
 814 The case of drainage of water-saturated medium sand at low DNAPL injection flow rate  
 815 ( $Q=29.6 \times 10^{-4}$  mL/min).  $t_{\text{mean}}$  is the average arrival time of the DNAPL front.

816 Fig. 5: The 3D view of the simulated DNAPL saturation field for vertical-downward drainage  
 817 of saturated medium-sized sand at low DNAPL injection flow rate using a 3D lattice network

818 (7×7×4858 nodes): (a) near the inlet, between z=0 cm and z=-1.5 cm; (b) in the middle of the  
819 soil column, between z= -34 cm and z=-35.5 cm; and (c) near the outlet, between z= -66.5 cm  
820 and z=-68 cm.

821 Fig. 6: Measured and simulated saturation as a function of depth at the end of the experiment  
822 using a 3D lattice network (20×20×200) with 3 different pore throat radius distributions: (a) in  
823 the case of vertical-upward drainage of water-saturated medium sand at low DNAPL injection  
824 flow rate and (b) in the case of vertical-downward drainage of water-saturated medium sand at  
825 low DNAPL injection flow rate.

826 Fig. 7: Effect of the size of the cross-section (6×6, 12×12, and 24×24) on the simulated DNAPL  
827 saturation field and cross-section averaged DNAPL saturation profile: in the case of vertical-  
828 downward drainage of saturated medium-sized sand at low DNAPL injection flow rate ( $Q=$   
829  $29.6 \times 10^{-4}$  mL/min), after 8 sec of DNAPL injection.

830 Fig. 8: Measured and simulated inlet DNAPL pressure as a function of time using a 3D network  
831 (7×7×4858): (a) in the case of vertical-upward drainage of water-saturated medium sand at low  
832 DNAPL injection flow rate and (b) in the case of vertical-downward drainage of water-saturated  
833 medium sand at low DNAPL injection flow rate ( $Q= 29.6 \times 10^{-4}$  mL/min).

834 Fig. 9: Measured and simulated dimensionless arrival times of the water/DNAPL front in  
835 medium sand. The case of vertical-downward drainage of water-saturated medium sand at high  
836 DNAPL injection flow rate ( $Q=59.2 \times 10^{-4}$  mL/min).  $t_{\text{mean}}$  is the average arrival time of the  
837 DNAPL front

838 Fig. 10: Measured and predicted DNAPL saturation as a function of depth at the end of the  
839 experiment using a 3D lattice network (20×20×200) with 3 different pore throat radius  
840 distributions. The case of vertical-downward drainage of water-saturated medium sand at high  
841 DNAPL injection flow rate ( $Q=59.2 \times 10^{-4}$  mL/min).

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