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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

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

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

1. Introduction

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

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

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

investigated to reproduce unstable displacement experiments. Kiriakidis et al. (1991) developed a 2D algorithm involving Monte Carlo decision-making, random walks, and principles of percolation theory. This algorithm successfully predicted the three distinct behaviours of immiscible displacement in porous media, such as viscous fingering, stable displacement, and invasion percolation, but the model did not include the gravity term, and gravity fingering was not considered. Ewing and Berkowitz (1998) developed a generalized growth model based on invasion percolation to simulate immiscible displacement in saturated porous media. Even capillary, viscous and gravity forces were incorporated in their 3D algorithm, where viscous forces were expressed stochastically rather than explicitly. Glass et al. (2001) developed a macro-modified invasion percolation (MMIP) model, including also the effects of all 3 forces within the invading phase in a macro-heterogeneous porous medium. The simulated evolving invasion structure during the gravity-destabilized condition matched the experimental distributions quite well. Dong et al. (2011) proposed a LBM model taking into account both wettability and gravity to successfully reproduce flow experiments in Hele Shaw cells. Recently, Liu et al. (2013; 2014; 2015) have developed a Lattice Boltzmann model that was able to simulate the main flow regimes (capillary fingering, viscous fingering and stable displacement). The model was applied to simulate drainage micromodel experiments of the liquid CO₂ displacement of water. The authors obtained a good agreement with the experimental measurements and showed that increasing M (viscosity ratio of non-wetting fluid to wetting fluid) or decreasing the media heterogeneity can enhance the stability of the displacement process.

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

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

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

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

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

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

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

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

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

2. Laboratory experiments on DNAPL migration

Laboratory experiments were conducted on a 68-cm long glass column with an internal diameter of 10 cm. The used porous medium is a medium-sized quartz sand with a mean grain-size diameter of 0.4 mm, a uniformity coefficient of 2.1 and a low fraction of organic content (foc = 0.09%, based on NFT 31–109) (Bohyet al. 2006). The hydraulic properties of the quartz sand, such as its moisture characteristics, were quantified in former studies by Cotel(2008). Trichloroethylene (TCE) was chosen as the DNAPL for the experiments because it is among the most frequently detected contaminants in subsurface environments. At 20 °C, TCE has a density of 1.463 g/cm³, a viscosity of 0.0056 g/s/cm, and a solubility in water of 1300 mg/L.

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

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

3. Numerical model

3.1. Geometry of the network

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

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

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

3.2. Governing flow equations

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

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

where $V_i [L^3]$ represents the volume of pore body i , $S_i^\alpha [-]$ represents local saturation (percent of V_i filled with fluid α), $Q_{ij}^\alpha [L^3 T^{-1}]$ is the phase flow rate through a pore throat connecting pore 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, unless the pore is located at the flow boundary. The sum runs over all neighbouring pores of pore body i .

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

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

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

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

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

where $\mu_\alpha [\text{ML}^{-1}\text{T}^{-1}]$ and $\rho_\alpha [\text{ML}^{-3}]$ are the dynamic viscosity and the density of the phase fluid present in the pore throat, respectively; r_{ij} and L_{ij} are the radius and the length of the pore throat, 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 vertices at the centre of pore bodies i and j . The first term of the right-hand side of Eq. (4), $\pi r_{ij}^4 / (8 L_{ij} \mu_\alpha)$, represents the flow conductance in the pore throat.

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

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

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

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

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

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

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

243 the following:

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

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:

$$251 \quad A \times B = P \tag{9}$$

252 where A is the conductance matrix with elements that depend on the connections between
253 different throats, P denotes the pressure vector, and the 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

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

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

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

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

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

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

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

3.4. Model parameters and numerical implementation

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

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

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

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

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

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

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

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

4. Results and discussion

4.1 Water retention curve modelling

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

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

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

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

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

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

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

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

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

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

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

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

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

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

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

4.2.2. DNAPL saturation

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

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

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

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

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

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

4.2.3. DNAPL pressure at the inlet section

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

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

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

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

4.2.4. Influence of the DNAPL injection rate on gravity fingering

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

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

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

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

5. Conclusions

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

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

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

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

Figure captions

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

Fig. 2: Experimental and modelled WRC of the quartz sand used in the laboratory experiments. Simulations with a 3D lattice network ($20 \times 20 \times 200$) and 3 different pore throat radii distributions. The capillary pressure value is expressed as equivalent water head (m H₂O).

Fig. 3: Experimental and modelled WRC of the quartz sand used in the laboratory experiments. Simulations with a 2D lattice network with 3 different sizes (50×50 , 75×75 , and 100×100) and a given pore-throat radii distribution ($r_{\max}-r_{\text{mean}}=1\text{sd}$). The capillary pressure value is expressed as equivalent water head (m H₂O).

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

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

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

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

Fig. 7: Effect of the size of the cross-section (6×6, 12×12, and 24×24) on the simulated DNAPL saturation field and cross-section averaged DNAPL saturation profile: in the case of vertical-downward drainage of saturated medium-sized sand at low DNAPL injection flow rate ($Q=29.6\times10^{-4}$ mL/min), after 8 sec of DNAPL injection.

Fig. 8: Measured and simulated inlet DNAPL pressure as a function of time using a 3D network (7×7×4858): (a) in the case of vertical-upward drainage of water-saturated medium sand at low DNAPL injection flow rate and (b) in the case of vertical-downward drainage of water-saturated medium sand at low DNAPL injection flow rate ($Q=29.6\times10^{-4}$ mL/min).

Fig. 9: Measured and simulated dimensionless arrival times of the water/DNAPL front in medium sand. The case of vertical-downward drainage of water-saturated medium sand at high DNAPL injection flow rate ($Q=59.2\times10^{-4}$ mL/min). t_{mean} is the average arrival time of the DNAPL front

Fig. 10: Measured and predicted DNAPL saturation as a function of depth at the end of the experiment using a 3D lattice network (20×20×200) with 3 different pore throat radius distributions. The case of vertical-downward drainage of water-saturated medium sand at high DNAPL injection flow rate ($Q=59.2\times10^{-4}$ mL/min).