Multifluid flow in bedded porous media: laboratory experiments and numerical simulations

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Understanding light nonaqueous-phase liquid (LNAPL) movement in heterogeneous vadose environments is important for effective remediation design. We investigated LNAPL movement near a sloping fine-over coarse-grained textural interface, forming a capillary barrier. LNAPL flow experiments were performed in a glass chamber (50 cm × 60 cm × 1.0 cm) using two silica sands (12/20 and 30/40 sieve sizes). Variable water saturations near the textural interface were generated by applying water uniformly to the sand surface at various flow rates. A model LNAPL (Soltrol 220) was subsequently released at two locations at the sand surface. Visible light transmission was used to quantitatively determine water saturations prior to LNAPL release and to observe LNAPL flow paths. Numerical simulations were performed using the Subsurface Transport Over Multiple Phases (STOMP) simulator, employing two nonhysteretic relative permeability–saturation–pressure (k–S–P) models. LNAPL movement strongly depended on the water saturation in the fine-grained sand layer above the textural interface. In general, reasonable agreement was found between observed and predicted water saturations near the textural interface and LNAPL flow paths. Discrepancies between predictions based on the van Genuchten/Mualem (VGM) and Brooks–Corey/Burdine (BCB) k–S–P models existed in the migration speed of the simulated LNAPL plume and the LNAPL flow patterns at high water saturation above the textural interface. In both instances, predictions based on the BCB model agreed better with experimental observations than predictions based on the VGM model. The results confirm the critical role water saturation plays in determining LNAPL movement in heterogeneous vadose zone environments and that accurate prediction of LNAPL flow paths depends on the careful selection of an appropriate k–S–P model. © 1998 Elsevier Science Limited. All rights reserved

Keywords: multifluid flow, LNAPL, capillary barrier, numerical simulation, constitutive relations.

1 INTRODUCTION

Light nonaqueous-phase liquids (LNAPLs) are common contaminants in soil and groundwater systems. Petroleum products spilled at the soil surface, and/or leakages from underground fuel storage facilities followed by vertical product migration through the vadose zone, are common LNAPL sources. While many investigations over the past decade have focused on the movement of LNAPLs in homogeneous porous media (e.g. Eckberg and Sunada,7 Abdul,1 Lenhard et al.,22,23 Cary et al.,4,5 Höst-Madsen and Jensen,13 Ostendorf et al.,46 Van Geel and Sykes,54 and Schroth et al.,45), the understanding of LNAPL behavior in heterogeneous subsurface environments is still incomplete. Tools have been developed to predict the migration of LNAPLs through the subsurface, but their evaluation is currently hindered by the limited availability of quantitative experimental data, especially in heterogeneous porous media.

Commonly encountered forms of geological heterogeneity
in alluvial depositional environments are alternating layers of fine- and coarse-grained porous media. In the vadose zone, a capillary barrier may form when a fine-grained layer overlays a coarse-grained layer and the interface between the two layers, i.e. the textural interface, is inclined. At a capillary barrier, percolating water is held in the fine-grained layer by capillary forces and is diverted downgradient, parallel to the sloping textural interface. The important role of capillary barriers in water flow and solute transport in the vadose zone has been demonstrated in laboratory experiments (e.g. Miyazaki\textsuperscript{29} and Steenhuis \textit{et al.}\textsuperscript{24}), field experiments\textsuperscript{8,19} and by numerical simulations.\textsuperscript{32} Predictive expressions describing water flow near a capillary barrier were given by Ross,\textsuperscript{42} and were later amended and extended by Steenhuis \textit{et al.}\textsuperscript{23,24} Stormont\textsuperscript{47} and Selker.\textsuperscript{47}

Effects of geological heterogeneity on nonaqueous-phase liquid (NAPL) migration at the field scale were investigated by Osborne and Sykes,\textsuperscript{35} who concluded that the extent of dense nonaqueous-phase liquid (DNAPL) migration was very sensitive to heterogeneities and anisotropies of porous media hydraulic properties. Similar results were obtained numerically by Kueper and Frind,\textsuperscript{47} and experimentally by Poulsen and Kueper.\textsuperscript{47} Essaid \textit{et al.}\textsuperscript{8} investigated a crude oil spill site and also concluded that the oil saturation distribution appeared, at least in part, to be influenced by heterogeneities in porous media hydraulic properties.

Controlled laboratory experiments of LNAPL infiltration and redistribution in layered porous media were conducted by several investigators. Cary \textit{et al.}\textsuperscript{13} performed experiments in one-dimensional vertical columns packed with horizontal layers of soils of different texture. The authors observed effects of layering on water and LNAPL movement, and used this information to partially verify a predictive model for LNAPL flow. Similarly, Oostrom and Lenhard\textsuperscript{34} conducted LNAPL infiltration and redistribution experiments in a vertical column packed with both homogeneous and heterogeneous (layered) porous media to compare parametric models used in simulating LNAPL subsurface migration. Pantazidou and Sitar\textsuperscript{45} conducted two-dimensional LNAPL infiltration experiments in layered sands and found that LNAPL movement was strongly dependent on the number and lateral continuity of layers and that, in general, more spreading of LNAPL occurred in layered than in homogeneous sand packs. Recently, Schroth \textit{et al.}\textsuperscript{46} conducted two-dimensional experiments to investigate LNAPL migration in the vicinity of textural interfaces. They found that LNAPL flow patterns in the vicinity of textural interfaces strongly depended on the water saturation at those locations and that LNAPL and water flow paths were mostly divergent. However, their experimental analysis was limited to one water flow rate and thus the observed LNAPL flow patterns were not investigated for a wider range of water saturations in the vicinity of the textural interface. In addition, water saturations near the textural interface were not measured quantitatively.

Numerous recent studies have focused on numerical simulations of multifluid flow phenomena (e.g. Forsyth\textsuperscript{10,12} Falta \textit{et al.}\textsuperscript{9,10} Sleep and Sykes\textsuperscript{46,49} Kaluarachchi and Parker,\textsuperscript{14} Mendoza and Frind,\textsuperscript{27} and Mendoza and McAlary\textsuperscript{36}). A critical component of multifluid flow simulations is the representation of the constitutive three-fluid relative permeability–saturation–pressure (\textit{k}–\textit{S}–\textit{P}) relationships. Parker \textit{et al.}\textsuperscript{29} developed a constitutive \textit{k}–\textit{S}–\textit{P} model for three-fluid air–NAPL–water systems based upon the two-fluid air–water van Genuchten/Mualem (VGM) \textit{k}–\textit{S}–\textit{P} model.\textsuperscript{55} White \textit{et al.}\textsuperscript{58} developed a numerical simulator (Subsurface Transport Over Multiple Phases, STOMP) which incorporates the \textit{k}–\textit{S}–\textit{P} model of Parker \textit{et al.}\textsuperscript{29} along with two other three-fluid \textit{k}–\textit{S}–\textit{P} models, one based on the two-fluid \textit{S}–\textit{P} model of Brooks and Corey\textsuperscript{3} and the pore size distribution model of Burdine\textsuperscript{3} (BCB), and the second being a hysteretic version of the VGM model (HVGM).\textsuperscript{20,25,38} Lenhard \textit{et al.}\textsuperscript{24} successfully conducted verification and validation exercises for STOMP for multifluid flow in a homogeneous sand pack, finding that STOMP provided accurate predictions of multifluid flow phenomena upon proper calibration of model parameters. Recently, Oostrom and Lenhard\textsuperscript{34} used STOMP to compare the accuracy of multifluid flow predictions based on the VGM, BCB and HVGM models. They compared simulator predictions with quantitative \textit{S}–\textit{P} data from one-dimensional LNAPL infiltration and redistribution experiments and found better agreement when employing the BCB model rather than the HVGM or VGM models. Most recently, Oostrom \textit{et al.}\textsuperscript{33} used STOMP to simulate LNAPL infiltration and redistribution in a two-dimensional homogeneous sand pack and found reasonable agreement between experimental and numerical data. So far, however, STOMP and the incorporated constitutive \textit{k}–\textit{S}–\textit{P} models have not been tested for other than one-dimensional problems of multifluid flow in heterogeneous (layered) porous media.

The experimental objectives of this study were to extend the work of Schroth \textit{et al.}\textsuperscript{46} to observe LNAPL movement near a textural interface for a variety of water saturations in the fine-grained layer above the interface. In addition, we wanted to obtain quantitative information on water saturations near the textural interface. Numerical simulation objectives were to investigate the predictability of multifluid flow in the vicinity of textural interfaces by comparing STOMP simulator predictions with experimental observations. Finally, we wanted to evaluate the effect that different constitutive \textit{k}–\textit{S}–\textit{P} models have on the predictive accuracy of multifluid flow simulations in the vicinity of textural interfaces.

2 MATERIALS AND METHODS

2.1 Overview

Transient two-dimensional LNAPL flow experiments were performed in a glass chamber designed to simulate
Multifluid flow near a sloping textural interface. The interface was constructed by packing a sloping layer of coarse-grained sand within a fine-grained sand. Experiments were conducted using different water flow rates to generate variable water saturation conditions in the fine-grained sand above the textural interface. In each experiment, LNAPL was released at two locations at the sand surface. Visible light transmission was used to quantitatively measure water saturations prior to LNAPL release and to observe LNAPL and water movement in the vicinity of the textural interface.

Measured water saturations and water and LNAPL flow paths were compared to predicted saturations and flow paths from numerical simulations. Simulations were conducted using both nonhysteretic VGM and nonhysteretic BCB $k$–$S$–$P$ models.

2.2 Porous media and fluid properties

Experiments were conducted using two grades of silica sand (Accusand® 12/20 (coarse-grained) and 30/40 (fine-grained) sieve sizes), manufactured by Unimin Corp. (Le Sueur, MN, U.S.A.). The sands were obtained prewashed (with water) and presieved by the manufacturer. Both sands feature high uniformity ($d_{60}/d_{10} = 1.23$ for the coarse-grained sand and 1.21 for the fine-grained sand), high sphericity (0.9 for both sands), high chemical purity and very low organic matter (< 0.04%). Main drainage water retention functions (Fig. 1) were obtained from three-fluid air–NAPL–water retention measurements using the method of Lenhard and by fitting the van Genuchten and Brooks–Corey models to the experimental data using a nonlinear least squares routine. The saturated hydraulic conductivity was 30.19 cm min$^{-1}$ for the coarse-grained sand and 8.94 cm min$^{-1}$ for the fine-grained sand, as measured by Schroth et al., using the constant-head method.

The model LNAPL selected for this study was Soltrol® 220 (Phillips Petroleum Co., Bartlesville, OK, U.S.A.), hereafter referred to as Soltrol. Alternative Soltrol formulations have been widely used to study LNAPL behavior in laboratory experiments (e.g. Brooks and Corey, Lenhard, Cary et al., and Oostrom and Lenhard). Soltrol is a mixture of branched $C_{12}$–$C_{17}$ alkanes that occur in a variety of petrochemical products, e.g. diesel fuel. Soltrol has a specific gravity of 0.81, a viscosity of 0.0047 Pa s, a negligible solubility in water, a low volatility at room temperature and a low health hazard. The aqueous phase in our experiments was distilled water.

2.3 Two-dimensional LNAPL flow experiments

Multifluid flow experiments were carried out in a chamber consisting of two 1.27-cm-thick glass panels (50 cm wide × 60 cm high), separated by 1.0-cm-thick aluminum spacers with inset rubber gaskets (Fig. 2). The textural interface was generated by packing a sloping layer of coarse-grained sand of 5.0 cm vertical thickness into the chamber, which was otherwise packed with fine-grained sand. A detailed description of the packing method is given in Schroth et al. In all experiments, the textural interface had a slope of 21.8° (4 vertical : 10 horizontal), was located 17 cm below the sand surface at the left edge of the sand pack, and had its downdip limit (physical end) a horizontal distance of 5.0 cm from the right edge of the sand pack (Fig. 2). The average porosity of the entire sand pack (combined coarse- and fine-grained sand) for all experiments, calculated from the total mass of each sand pack and the packing height, was 0.35 with a standard deviation of 0.01. Some packing variability was visible in the slightly wavy appearance of the capillary fringe, likely caused by porosity variations due to wall effects. However, its effect on the LNAPL flow experiments was negligible, as verified by the consistent results of experiments performed with replicate packings in the same experimental chamber.

In all experiments, the sand-packed chamber was first purged of air by injecting CO$_2$ gas upward through the manifold in the bottom of the chamber (Fig. 2). Distilled water was then pumped from the bottom manifold into the sand pack until the sand pack was fully water saturated. Visual observation confirmed no gas bubbles in the sand pack following this procedure. Once the sand pack was water saturated, a water irrigation manifold containing 10 equally-spaced syringe needles was placed on top of the chamber, and water was applied to the sand surface throughout the remainder of the experiments at constant flow rates ranging between 2.0 and 28.9 cm$^3$ min$^{-1}$ using a piston pump. During water application, the overflow reservoir was lowered to establish a water table at the bottom of the chamber above the manifold and was maintained for at least 12 h prior to LNAPL release. A 12.0-cm$^3$ pulse of Soltrol was then released at location B at the sand surface (Fig. 2), at a flow rate of 0.1 ± 0.01 cm$^3$ min$^{-1}$, using a syringe needle connected to a peristaltic pump. A similar Soltrol pulse was released at location A approximately 24 h after the release of Soltrol at location B. The experiments were...
terminated once the plume of LNAPL released at location A reached the zone of migration of the plume released at location B in the vicinity of the textural interface. Air within the sand pack was maintained at atmospheric pressure throughout the duration of the experiments by allowing air movement through the top of the experimental chamber. After each experiment, the experimental chamber was disassembled, cleaned and repacked with clean sand.

2.4 Water/LNAPL flow visualization and water saturation measurements

Visible light transmission was used to measure water saturations prior to LNAPL release and to delineate water and LNAPL flow paths during and after LNAPL release. Equally spaced fluorescent lights were mounted behind the experimental chamber and the light intensity transmitted through the sand pack was recorded by a charge-coupled device (CCD) color camera focused on the front glass panel. The camera output was digitized into 24-bit (eight bits each for red, green and blue light), 640 × 480 pixel images and stored for subsequent analysis. For each experiment, images of transmitted light intensity were recorded for air-dry and water-saturated conditions, at steady-state water flow prior to LNAPL release, and during and after LNAPL release. The ambient room temperature was kept constant at 23°C throughout the experiments. No increase in fluid temperatures due to heat generated by the light source was measured on the front glass panel.

To identify whether water was fully diverted along the textural interface prior to the release of LNAPL, the movement of a blue dye (FD&C Blue No. 1, Crompton and Knowles, Reading, PA, U.S.A.) was visually observed by releasing approximately 1 cm³ of dye solution at the left corner of the chamber at the sand surface. The dye was flushed out of the sand pack prior to the release of NAPL. LNAPL flow paths were obtained by visually tracing the outline of the Soltrol plume from the recorded images. To improve the delineation between water and Soltrol by light transmission, Soltrol was dyed with 0.01% by weight of Sudan III (Matheson, Coleman and Bell, Manufacturing Chemists, Norwood, OH, U.S.A.), a hydrophobic stain of bright red color which is virtually insoluble in water.

Water saturations at steady-state water flow prior to the release of LNAPL were computed from the recorded images using a modified version of the method of Tidwell and Glass. They derived an expression for the predicted water saturation, \( S_w \), as a function of the normalized transmitted light intensity, \( I_w \):

\[
S_w = \frac{\ln \left( I_w \left( \tau_{sw} / \tau_{sa} \right)^{3/2} - 1 \right) + 1}{2 \ln \left( \tau_{sw} / \tau_{sa} \right)}
\]

where \( \tau_{sw} \) and \( \tau_{sa} \) are light transmission factors for the sand–water and sand–air interfaces, calculated to be

![Fig. 2. Experimental chamber used for conducting transient two-dimensional LNAPL flow experiments.](image)
where \( I_w \) is the transmitted light intensity for which the water saturation is desired, \( I_d \) and \( I_s \) are the transmitted light intensities for air-dry and water-saturated conditions, respectively. Therefore, eqn (1) predicts water saturations ranging from air-dry to fully saturated conditions \( (I_d \leq I_s \leq I_w) \). In our LNAPL flow experiments (performed in water-wetted porous media), however, the smallest transmitted light intensity observed was \( I_v \), which occurred when the porous medium was at the irreducible water saturation, \( S_{irr} \). We modified eqn (1) to reflect these specific experimental conditions, to yield:

\[
S_{irr} = \frac{\ln\{a(I_d - I_w)(I_w/I_s)^{a} - 1 + 1\}}{2f \ln(I_w/I_s)} + S_m
\]

where \( a \) is an additional fitting parameter.

The parameters \( a, I_d \) and \( f \) in eqn (3) are porous medium dependent and therefore require a separate calibration for each sand grade (as does \( f \) in eqn (1)). Two additional experiments were conducted for this purpose, in which the experimental chamber was packed exclusively with either coarse-grained or fine-grained sand. Following the procedure described in the previous section, the air-dry sand pack was fully water saturated. By lowering the overflow reservoir, water was then allowed to drain from the sand pack for a least 24 h, after which all outflow from the chamber had ceased and hydrostatic conditions were assumed. Transmitted light intensities were recorded for air-dry, water-saturated (Table 1) and hydrostatic conditions. Normalized light intensities for the hydrostatic conditions were computed (from the red light) for each image pixel using eqn (2). Horizontal averages of \( I_n \) were then obtained for the interval of \( 5 \) cm around the horizontal center of the experimental chamber. These were used in eqn (3) to obtain best fits of \( a, I_d \) and \( f \) (Fig. 3, Table 1) by minimizing

\[
\sum (S_{irr} - S_{irr,VG})^2
\]

using a nonlinear least squares routine, where \( S_{irr,VG} \) is the hydrostatic water saturation predicted by the van Genuchten \(^{55} \) retention function, which was determined for the sand grades utilized in this study by Schroth et al. \(^{44} \)

### Table 1. Recorded light intensities obtained during parameter calibration and best fit parameters for quantitative determination of water saturation as a function of normalized transmitted light intensity (eqn (3)) for two sand grades

<table>
<thead>
<tr>
<th>Accusand (^{®} ) grade</th>
<th>Air-dry condition</th>
<th>Saturated condition</th>
<th>( a )</th>
<th>( I_d )</th>
<th>( f )</th>
<th>( S_m )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coarse (12/20)</td>
<td>64.7 ± 9.82</td>
<td>197.3 ± 25.6</td>
<td>1.44</td>
<td>0.31</td>
<td>1.34</td>
<td>0.034</td>
</tr>
<tr>
<td>Fine (30/40)</td>
<td>38.7 ± 3.8</td>
<td>206.0 ± 25.8</td>
<td>1.25</td>
<td>0.25</td>
<td>7.87</td>
<td>0.052</td>
</tr>
</tbody>
</table>

\(^{†}\)Not fitted, determined independently from water retention measurements. \(^{44} \)

\(^{‡}\)Average ± standard deviation for entire sand pack, obtained from the eight-bit red light (range 0–255).

### 2.5 Numerical simulations

Numerical simulations were conducted using the STOMP simulator. \(^{56,57} \) STOMP, an acronym for Subsurface Transport Over Multiple Phases, is a three-dimensional, three-fluid compositional simulator for modeling contaminant flow and transport in variably saturated geological media. \(^{31} \) Simulations of our laboratory experiments were conducted with the nonvolatile three-fluid operational mode (NVTP). \(^{58} \) This mode solves isothermal problems involving the simultaneous flow of an aqueous phase and an NAPL with sufficiently small vapor pressure (such as Soltrol) that NAPL transport through the gas phase can be neglected and constant air pressure can be assumed. Under the additional assumptions of negligible mutual liquid solubility and negligible NAPL adsorption onto the porous media, the governing mass conservation equations for water (subscript w) and NAPL (subscript o) components are

\[
\frac{\partial}{\partial t} [\rho_w S_w] = - [\nabla \cdot [\rho_w q_w]] + m_w
\]

\[
\frac{\partial}{\partial t} [\rho_o S_o] = - [\nabla \cdot [\rho_o q_o]] + m_o
\]

![Fig. 3. Calibration of normalized transmitted light intensity, \( I_n \), to water saturation, \( S_{irr} \), for coarse-grained (12/20) sand under hydrostatic conditions. The best fits of eqns (1) and (3) (predicted water saturations, \( S_w \)) to water saturations obtained from the coarse-grained sand van Genuchten (VG) water retention function are shown.](image-url)
with
\[
\mathbf{q}_w = -\frac{k_i}{\rho_w} \nabla P_w + \rho_w \mathbf{g} z
\]
and
\[
\mathbf{q}_a = -\frac{k_i}{\rho_a} \nabla P_a + \rho_a \mathbf{g} z
\]
where \( t \) is time, \( \eta \) is effective porosity, \( \rho \) is density, \( S \) is fluid saturation, \( \mathbf{q} \) is the volumetric flux vector, \( m \) is the component mass source rate, \( k \) is the intrinsic permeability tensor, \( k_i \) is relative permeability, \( \mu \) is viscosity, \( P \) is pressure, \( g \) is the gravitational constant and \( z \) is the vector in the direction of gravitation.

In STOMP, the governing equations are discretized using the integrand-volume finite difference method\(^{30}\) with Euler backward (fully implicit) time differencing, and are solved for the primary variables \( P_w \) and \( P_a \) with a multi-variable, residual-based Newton–Raphson iteration technique.\(^{16}\) Secondary variables (e.g. \( S_w, S_a \) etc.) are computed from the primary data set through the constitutive relations. Non-hysteretic VGM and BCB \( k-S \)–\( P \) models were used for the simulations presented here. Assuming fluid wettability increases in the order air < NAPL < water, and that NAPL completely wets water surfaces, the total liquid saturation \( S_l \)
\[
S_l = S_w + S_n
\]
will be a function of the air–NAPL capillary head, and \( S_n \) will be a function of the NAPL–water capillary head.\(^{38}\) In the VGM model, \( S-P \) relations are based on the van Genuchten\(^{33}\) retention function, where the effective water saturation \( S_w \)
\[
S_w = \frac{S_w - S_n}{1 - S_n}
\]
in a three-fluid system is given by\(^{39}\)
\[
\tilde{S}_w = [1 + (a \beta_n h_{wo})^\gamma]^{-n} \quad h_{wo} > 0
\]
\[
\tilde{S}_w = 1 \quad h_{wo} \leq 0
\]
and the effective total liquid saturation \( \tilde{S}_l \)
\[
\tilde{S}_l = \frac{S_l - S_n}{1 - S_n}
\]
is given by
\[
\tilde{S}_l = [1 + (a \beta_n h_{wo})^\gamma]^{-n} \quad h_{wo} > 0
\]
\[
\tilde{S}_l = 1 \quad h_{wo} \leq 0
\]
where \( a \) and \( n \) are van Genuchten retention parameters, \( m = 1 - 1/n \), \( h_{wo} \) is the water-equivalent NAPL–water capillary head with \( h_{wo} = (P_w - P_a) / \rho \), \( h_{wo} \) is the water-equivalent air–NAPL capillary head with \( h_{wo} = (P_w - P_a) / \rho P_a \), \( P_a \) is air pressure, and \( \beta_n \) and \( \beta_{wo} \) are interfacial tension-dependent scaling factors.\(^{21}\) In the VGM model, water and NAPL relative permeabilities, \( k_w \) and \( k_n \) are derived from Mualem’s\(^{30}\) pore size distribution model and are given by\(^{20}\)
\[
k_w = \frac{S_w^{1/2} \left[ 1 - (1 - S_w)^m \right]^{1/2}}{S_w}
\]
\[
k_n = \frac{(S_l - S_n)^{1/2} \left[ (1 - S_w)^m - (1 - S_n)^m \right]^{1/2}}{S_l - S_n}
\]
In the BCB model, \( S-P \) relations are based on the Brooks and Corey\(^{22}\) retention function, and \( \tilde{S}_w \) and \( \tilde{S}_n \) in a three-fluid system are given by\(^{34}\)
\[
\tilde{S}_w = \left( \frac{h_d}{\beta_n h_{wo}} \right)^\lambda \beta_n h_{wo} \leq h_d
\]
\[
\tilde{S}_n = 1 \quad \beta_n h_{wo} > h_d
\]
\[
\tilde{S}_n = \left[ \frac{h_d}{\beta_n h_{wo}} \right]^\lambda \beta_n h_{wo} \leq h_d
\]
\[
\tilde{S}_l = 1 \quad \beta_n h_{wo} > h_d
\]
where \( h_d \) is the water-equivalent air-entry (displacement) capillary head and \( \lambda \) is a pore size distribution factor. In the BCB model, \( k_w \) and \( k_n \) are derived from Burdine’s\(^{5}\) pore size distribution model and are given by\(^{34}\)
\[
k_w = \frac{S_w^{2+1/\lambda}}{S_w}
\]
\[
k_n = \left( \frac{S_l - S_n}{S_l} \right)^{2+1/\lambda}
\]
The problem domain was discretized using a two-dimensional grid that contained 1505 nodes (35 \( \times \) 43) and was tilted to align one coordinate direction with the textural interface (Fig. 4). A tilted gravitational vector was used to compensate for this alignment. The grid spacing was 2.0 cm in the \( x \)-direction and varied between 0.5 and 4.0 cm in the \( y \)-direction, with the smallest grid spacing just above the textural interface. Due to the tilting of the grid, some grid nodes (located at the geometric center of grid blocks; not shown) were located outside the experimental domain and were excluded from the computations, thus reducing the grid to 964 active nodes (Fig. 4). Porous media and fluid-related input parameters (Table 2) were determined in independent experiments reported by Schroth et al.\(^{44}\)

A hydrostatic condition, with the water table simulated slightly above the bottom of the computational domain, was chosen as the initial condition for all simulations. During the simulations, the water table was maintained at that location by setting a Dirichlet boundary condition along the bottom of the computational domain. Constant water application was simulated using a Neumann boundary condition for all nodes along the top of the computational domain. Once the simulations approached steady-state for the water phase (constant \( S_w \) throughout the computational
domain), the release of LNAPL at locations A and B was simulated using time-dependent Neumann boundary conditions at two nodes (Fig. 4), and the experimental LNAPL flow rate and pulse duration. At the bottom of the computational domain, a no-flow boundary for LNAPL was used. The side boundaries (left and right, Fig. 4) were modeled as no-flow boundaries for water and LNAPL.

For all simulations, the convergence tolerance for the pressures was $10^{-6}$ Pa and the maximum number of Newton–Raphson iterations per time step was 16. A time-step increment factor of 1.25 was used to increase time-step size to a maximum of 600 s prior to the simulated LNAPL release and to 60 s after the release. Upwind interfacial averaging was applied to the relative permeabilities. Harmonic averages were applied to the remaining flux components.

3 RESULTS AND DISCUSSION

3.1 Two-dimensional LNAPL flow experiments

Significant variations in water saturation, $S_w$, were observed under steady-state water flow conditions at locations A' and B' in the fine-grained sand just above the textural interface (Fig. 2) as a function of the water flux applied to the sand surface, shown in Table 3 as relative water flux, $q_w/K_{sat}$, where $q_w$ is the water flux applied per unit surface area and $K_{sat}$ is the saturated hydraulic conductivity of the fine-grained sand. In all experiments, $S_w$ was larger at B' than at A' due to water being diverted downgradient, parallel to the textural interface. In experiments 1–5, water was fully diverted parallel to the textural interface prior to LNAPL release, i.e. water did not penetrate the coarse-grained sand layer. In experiments 6 and 7, water partially penetrated the coarse-grained sand layer near the textural interface down dip limit prior to the LNAPL release (not shown).

It is important to note that the $S_w$ values in Table 3 were measured prior to LNAPL release. As such, these values may not reflect exact $S_w$ conditions present during LNAPL infiltration, when air–water interfaces were replaced by air–NAPL and NAPL–water interfaces, accompanied by a change in interfacial tension at the fluid–water interface. However, no quantitative measurement of $S_w$ was performed in the presence of LNAPL, because the measurement of fluid saturations by the light transmission technique has not yet been extended to three-fluid air–NAPL–water systems.

LNAPL flow patterns in the vicinity of the textural interface strongly depended on $S_w$ (Table 3). Full diversion of the migrating LNAPL plume along the interface was observed near location A' in experiment 1, with $0.35 < S_w < 0.45$ (Fig. 5(a)). This may be explained by considering the distribution of fluids within the pore space. Fluid wettability in water-wet porous media increases in the order air, LNAPL, water, i.e. in the presence of all three fluids water will occupy the smallest pores, LNAPL will occupy

![Fig. 4. Finite difference grid used in simulations of LNAPL flow experiments. Nodes (not drawn) are located at grid block centers. Bold black contour lines show the chamber boundaries and the location of the coarse-grained sand layer. Shaded grid blocks represent nodes excluded from the computations.](image)

Table 2. Porous media and fluid-related STOMP simulator input parameters determined in independent experiments reported by Schroth et al.44

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Sand grade</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Coarse (12/20)</td>
</tr>
<tr>
<td>van Genuchten $\alpha$</td>
<td>0.151</td>
</tr>
<tr>
<td>van Genuchten $n$</td>
<td>7.35</td>
</tr>
<tr>
<td>Brooks and Corey $h_3$</td>
<td>5.42</td>
</tr>
<tr>
<td>Brooks and Corey $\lambda$</td>
<td>3.94</td>
</tr>
<tr>
<td>Irreducible water saturation, $S_m$</td>
<td>0.034</td>
</tr>
<tr>
<td>Air–water scaling factor, $\beta_{aw}$</td>
<td>1.0</td>
</tr>
<tr>
<td>Air–NAPL scaling factor, $\beta_{aw}$†</td>
<td>2.38</td>
</tr>
<tr>
<td>NAPL–water scaling factor, $\beta_{wm}$†</td>
<td>1.76</td>
</tr>
<tr>
<td>Intrinsic permeability, $k$</td>
<td>$5.14 \times 10^{-6}$</td>
</tr>
<tr>
<td>Effective porosity, $\eta$</td>
<td>0.35</td>
</tr>
</tbody>
</table>

†NAPL dyed with 0.01% by weight of Sudan® III.
Table 3. Results of two-dimensional LNAPL flow experiments and simulator predictions of water saturations at locations A’ and B’ under steady-state water flow conditions using the van Genuchten/Mualem (VGM) and Brooks–Corey/Burdine (BCB) k–S–P models

<table>
<thead>
<tr>
<th>Experiment no.</th>
<th>Relative water flux $q_w / K_{s,fine}$</th>
<th>LNAPL release at A</th>
<th>Predicted $S_w$ at A’</th>
<th>Predicted $S_w$ at B’</th>
<th>LNAPL release at B</th>
<th>Predicted $S_w$ at B’</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Water saturation $S_w$ (at A’)</td>
<td>LNAPL penetration observed†</td>
<td>Water saturation $S_w$ (at B’)</td>
<td>LNAPL penetration observed†</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>0.0056</td>
<td>0.35–0.45 no</td>
<td>0.50–0.60 yes</td>
<td>0.36–0.41 0.40–0.45 0.58–0.59 0.59–0.61</td>
<td></td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>0.011</td>
<td>0.45–0.55 minor§</td>
<td>0.70–0.80 yes</td>
<td>0.49–0.58 0.53–0.60 0.76–0.77 0.80–0.82</td>
<td></td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0.017</td>
<td>0.60–0.70 yes</td>
<td>0.87–0.93 yes</td>
<td>0.57–0.66 0.59–0.68 0.85 0.92–0.95</td>
<td></td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>0.022</td>
<td>0.60–0.75 yes</td>
<td>0.90–0.95 minor§</td>
<td>0.63–0.73 0.64–0.74 0.90 0.99</td>
<td></td>
<td></td>
</tr>
<tr>
<td>5</td>
<td>0.028</td>
<td>0.70–0.85 yes</td>
<td>0.90–0.96 no</td>
<td>0.69–0.79 0.69–0.80 0.93 1.00</td>
<td></td>
<td></td>
</tr>
<tr>
<td>6</td>
<td>0.042</td>
<td>0.80–0.90 yes</td>
<td>—</td>
<td>0.80–0.89 0.81–0.96 — —</td>
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</tr>
<tr>
<td>7</td>
<td>0.081</td>
<td>0.95–0.98 no</td>
<td>—</td>
<td>0.96–0.98 1.00 — —</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

†Water saturation in fine-grained (30/40) sand just above the textural interface at the horizontal position (±2.5 cm) where LNAPL plume was released (measured/simulated prior to LNAPL release).
‡Partial penetration of LNAPL into coarse-grained (12/20) sand layer.
§Penetration of LNAPL as few isolated ganglia.
¶No LNAPL released, water penetration into coarse-grained sand layer observed during steady-state water flow.

Fig. 5. Location of migrating LNAPL plumes during flow experiments conducted at relative water flux conditions of: (a) 0.0056 (experiment 1) and (b) 0.028 (experiment 5). Plume boundaries are shown for elapsed times (h: min) since the start of LNAPL release.

The dotted line represents the contour of the zone of higher water saturation above the textural interface.
intermediate-size pores, and the largest pores will be air filled. The air–LNAPL capillary head, $h_{ao}$, is determined by the location of the air–NAPL interfaces within the pore space and by the air–LNAPL interfacial tension. The location of the air–LNAPL interfaces is a function of the total liquid saturation $S_t$, and hence is a function of $S_w$ (eqn (9)). For small $S_w$, $h_{ao}$ in the fine-grained sand was large enough to prevent LNAPL penetration into the coarse-grained sand. Since $h_{ao}$ increases with decreasing $S_t$, full diversion of LNAPL along the textural interface is expected for any $S_w \leq 0.45$ (Table 3) under otherwise identical experimental conditions. Similar observations of full LNAPL diversion along a textural interface for $S_w \leq 0.10$ were made by Schroth et al.\textsuperscript{46} Note that $S_t$ and thus $h_{ao}$ is also a function of $S_o$ (eqn (9)). Therefore, while not investigated in our experiments, the LNAPL flow pattern must also depend on the LNAPL release rate.

Partial penetration of LNAPL was observed at B’ in experiment 1 (Fig. 5(a)), at A’ and B’ in experiments 2–4, and at A’ in experiments 5 and 6 (Fig. 5(b)). In all these cases, $S_w$ in the fine-grained sand immediately above the textural interface ranged from 0.45 to 0.90. Partial penetration of LNAPL in the form of a few isolated ganglia was observed for even higher $S_w$ (up to 0.95; Table 3). In our experiments, LNAPL flow patterns appeared to be independent of the release location and to depend only on $S_w$ near the textural interface. Consequently, the location of partial penetration shifted upgradient along the textural interface with increasing water flux. Using similar reasoning as before, partial penetration of LNAPL into the coarse-grained sand layer may be explained by the reduction in $h_{ao}$ in the vicinity of the textural interface due to the increase in $S_w$ and thus $S_t$, allowing LNAPL to penetrate the coarse-grained sand layer.\textsuperscript{46}

Full diversion of LNAPL parallel to the textural interface occurred for $S_w \geq 0.90$ (Table 3). In these cases, however, LNAPL was diverted above the textural interface, supported on a zone where water occupied most of the pore space (Fig. 5(b)). Water acted as a barrier to LNAPL flow in similar fashion as it does during the formation of LNAPL lenses in static capillary fringes (e.g. Abdul,\textsuperscript{1} Schroth et al.,\textsuperscript{15} and Oostrom and Lenhard\textsuperscript{34}). Full diversion of LNAPL above the textural interface was observed at both B’ (experiment 5) and A’ (experiment 7; Table 3), providing further indication that $S_w$ strongly influences LNAPL flow in the vicinity of textural interfaces.

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**Fig. 5.** (Continued)
3.2 Numerical simulations

Predicted values of $S_a$ at $A'$ and $B'$ for steady-state water flow conditions prior to the release of LNAPL agreed well with measured values of $S_a$ at those locations (Table 3). In general, values of $S_a$ predicted using the VGM model were smaller than those predicted using the BCB model. Discrepancies between the two models were largest for very high $S_a$, with predicted values of $S_a$ approaching unity based on the BCB model, whereas predicted values based on the VGM model were almost 10% smaller (Table 3). This difference may, in part, be explained by considering the van Genuchten and Brooks–Corey water retention functions for the fine-grained sand (Fig. 1). For water-equivalent air–water capillary head, $h_{w}$, values smaller than 14 cm, predicted values of $S_a$ are generally smaller for the van Genuchten function than for the Brooks–Corey function. However, because the solution of the water flow equation depends on both $S$–$P$ and $k$–$S$ relations, the reasons for discrepancies between predictions based on the VGM and BCB models are more complex and difficult to assess due to the strong nonlinearities in these models.

Simulated steady-state water flow conditions for $q_{l}/K_{s,fine} = 0.0056$ (Fig. 6(a, b)) agreed well with experimental observations in that full diversion of water along the textural interface was predicted for simulations based on both VGM and BCB models. In addition, full diversion of the migrating LNAPL plume near $A'$ was predicted using both VGM and BCB models (Fig. 6(c, d)). No direct comparisons between experimental and predicted values of $S_a$ near $A'$, were possible due to the lack of quantitative experimental data. However, in agreement with qualitative experimental observations, predicted values of $S_a$ within the LNAPL plume near $A'$ were significantly larger than those predicted for locations well above the textural interface. An increase in $S_a$ along the textural interface was observed previously by Schroth et al.\(^{16}\) in continuous LNAPL release experiments and explained by the reduced gradient for LNAPL flow that occurred along the textural interface, which, combined with a reduction of cross-sectional area available to LNAPL flow, required an increase in $S_a$ to maintain steady-state LNAPL flow conditions.

A discrepancy between predictions based on the VGM and BCB models existed in the migration speed of the simulated LNAPL plume, shown in Fig. 6(c) and (d) at 2.5 h after the start of LNAPL release at location A. Simulations based on the VGM model predicted the tip of the LNAPL plume to move further downgradient along the textural interface than simulations based on the BCB model. Such differences in predicted migration speed may not only be due to differences in $S$–$P$ relations between the two models, but may also be due to differences in $k$–$S$ relations (eqns (14), (15), (18) and (19)). Oostrom and Lenhard\(^{34}\) showed that, for a fixed $S_w$, the ratio of relative permeabilities $k_{w} (VGM)/k_{w} (BCB)$ is $\approx 1$ for any $S_w$, which is in agreement with our simulation results. Furthermore, a comparison between the predicted position of the migrating LNAPL plume (Fig. 6(c, d)) and the experimentally observed position (Fig. 5(a)) shows better agreement for the prediction based on the BCB model, which agrees with the results obtained by Oostrom and Lenhard\(^{34}\) in one-dimensional column experiments and simulations.

Only a small fraction of LNAPL was predicted to penetrate the coarse-grained sand layer near $B'$ in simulations of LNAPL release at location B under steady-state water flow conditions ($q_{l}/K_{s,fine} = 0.0056$) (Fig. 6(c, f)). While comparisons could only be qualitative, it appeared that a larger fraction of LNAPL was observed to penetrate the coarse-grained sand layer during the LNAPL flow experiments (Fig. 5(a)) than was predicted. An explanation why simulator predictions disagreed with experimental observations may be given by noting that the effects of hysteresis in $k$–$S$–$P$ relationships were not included in the VGM and BCB models, although hysteretic conditions existed during the LNAPL flow experiments. Water was on the main drainage $S$–$P$ path throughout the experimental chamber until steady-state water flow conditions were established. However, during LNAPL infiltration, $S_w$ was on a primary imbibition $S$–$P$ path, followed by a secondary drainage $S$–$P$ path during LNAPL redistribution. As a consequence, $h_w$ was likely to be smaller in the experiments than predicted from nonhysteretic simulations based upon main drainage $S$–$P$ data, which could increase the LNAPL flux into the coarse-grained sand layer.

Simulated steady-state water flow conditions for $q_{l}/K_{s,fine} = 0.028$ (Fig. 7(a, b)) agreed well with experimental observations in that full diversion of water along the textural interface was predicted based on both VGM and BCB models. As discussed earlier, differences between VGM and BCB model predictions in $S_a$ were most pronounced for high $S_a$ near $B'$ (Table 3). In agreement with experimental observations (Fig. 5(b)), partial penetration of LNAPL into the coarse-grained sand layer was predicted near $A'$ for both VGM and BCB models (Fig. 7(c, d)). The VGM model predicted a somewhat larger fraction of LNAPL to penetrate the coarse-grained sand layer compared to the BCB model, but a conclusive comparison with experimental results was not possible, since LNAPL saturations were not determined quantitatively during the experiments.

Significant differences between predictions based on VGM and BCB models were observed for the simulated LNAPL plume near $B'$ under steady-state water flux conditions ($q_{l}/K_{s,fine} = 0.028$) (Fig. 7(e, f)). In disagreement with experimental observations, LNAPL penetration into the coarse-grained sand layer was predicted using the VGM model (Fig. 7(e)). On the other hand, predictions based on the BCB model showed excellent agreement with experimental observations in that the simulated LNAPL plume was fully supported on the zone of high $S_a$ above the textural interface and started to divert downgradient parallel to the textural interface (Fig. 7(f)). Differences between VGM and BCB model predictions were largely due to differences in the van Genuchten and...
Brooks–Corey $S–P$ relations. In simulations based on the
VGM model, the largest pores near $B'$ were air filled prior to
the release of LNAPL ($S_w = 0.93$; Table 3), thus providing a
flow path for LNAPL to the textural interface. In the simu-
lations based on the BCB model, water-filled pores (pre-
predicted $S_w = 1.0$ near $B'$ prior to the release of LNAPL;
Table 3) acted as a barrier to LNAPL flow, preventing
LNAPL from reaching the textural interface. To allow for
displacement of water from a water-saturated pore space in
the Brooks–Corey $S–P$ model, $P_o$ must exceed the critical
entry pressure, a value based on pore geometry. This is not
the case for the van Genuchten $S–P$ model, where LNAPL
will displace water from a water-saturated pore space if
$P_o > P_w$. Similar results were obtained by Oostrom and

Fig. 6. Simulator predictions for LNAPL flow experiment conducted at 0.0056 relative water flux; steady-state water saturation distri-
butions prior to the release of LNAPL using: (a) the van Genuchten/Mualem (VGM) and (b) the Brooks–Corey/Burdine (BCB) models.
LNAPL saturation distributions at 2.5 h after the start of LNAPL release at location A using the VGM (c) and the BCB (d) models, and at
6 h after the start of LNAPL release at location B using the VGM (e) and the BCB (f) models.
Lenhard, where LNAPL was predicted to penetrate into the water-saturated zone of a sandy medium in simulations based on the VGM model, but to accumulate in the upper zone of the capillary fringe in simulations based on the BCB model. Note that in our simulations based on the VGM model, water-saturated conditions would prevent full water diversion along the textural interface, because water breakthrough into the coarse-grained sand layer occurs when $h_{w} = 0$.

To further illustrate the discrepancy between predictions based on the VGM and BCB models, we consider the volumetric LNAPL fraction that was predicted to penetrate the coarse-grained sand layer as a function of the applied relative water flux at locations $A'$ and $B'$ (Fig. 8). Due to

**Fig. 7.** Simulator predictions for LNAPL flow experiment conducted at 0.028 relative water flux; steady-state water saturation distributions prior to the release of LNAPL using: (a) the van Genuchten/Mualem (VGM) and (b) the Brooks–Corey/Burdine (BCB) model. LNAPL saturation distributions at 2.5 h after the start of LNAPL release at location $A$ using the VGM (c) and the BCB (d) models, and at 6 h after the start of LNAPL release at location $B$ using the VGM (e) and the BCB (f) models.
the transient nature of the simulations, specific times (3 h after the start of the simulated LNAPL release for A′ and 6 h for B′) were chosen as a basis for comparison. These times were selected based on experimental observations that at these times most of the migrating LNAPL had reached the vicinity of the textural interface and had started to divert, at least in part, parallel to the interface. For both locations, discrepancies between VGM and BCB model predictions increased with increasing relative water flux, thus increasing $S_w$ just above the textural interface. While predictions based on the VGM model appeared to asymptotically approach a maximum for the penetrating LNAPL fraction with increasing relative water flux, predictions based on the BCB model initially showed an increase in penetrating LNAPL fraction, followed by a reduction to near zero with increasing relative water flux. The latter behavior was in much better agreement with experimental observations. Hence, more realistic predictions of LNAPL flow near textural interfaces appear to be obtained in simulations based on the BCB model, at least for the case of higher $S_w$ near the interfaces.

4 CONCLUSIONS

Experimental results confirmed the previous findings of Schroth et al.\textsuperscript{46} that the fate of LNAPL near textural interfaces is a strong function of the water saturation just above the interface. In agreement with our previous study, three distinct LNAPL flow patterns were identified for a range of water saturations. Full diversion of LNAPL above the textural interface was observed for $S_w \approx 0.45$, partial penetration of the migrating LNAPL plume into the coarse-grained sand layer was observed for $0.45 \leq S_w \leq 0.90$, and full diversion of LNAPL above the textural interface was observed for $S_w > 0.90$. Therefore, knowledge of water saturations during the migration of LNAPL plumes in layered subsurface systems is a prerequisite for predicting LNAPL migration patterns and thus environmental impacts of LNAPL on soil and groundwater.

In general, reasonable agreement was found between experimental observations and simulator predictions for $S_w$ above the textural interface under steady-state water flow conditions, as well as for most LNAPL flow patterns in the vicinity of the interface. Discrepancies between predictions based on the VGM and BCB models were noticed for the migration speed of the LNAPL plume, volume fractions of LNAPL that penetrated the coarse-grained sand for $0.45 \leq S_w \leq 0.90$, and most notably, for the predicted LNAPL flow patterns for $S_w > 0.90$.

Discrepancies between predictions by the VGM and BCB models were attributed to differences in the $S$–$P$ and $k$–$S$ relations used in the two models. Better agreement between experimental observations and the predicted migration speed of the LNAPL plume and LNAPL flow pattern for $S_w > 0.90$ was obtained when employing the BCB model. Hysteretic conditions in $S$–$P$ relations during the experiments were identified as a possible reason why predicted volume fractions of LNAPL penetrating the coarse-grained sand layer appeared to differ from experimental observations. However, no fully quantitative comparison was possible due to the current limitation of the visible light transmission method to one-liquid systems. More accurate predictions of LNAPL flow patterns near textural interfaces may be possible when employing hysteretic versions of $k$–$S$–$P$ models, in particular of the BCB model.

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