Numerical Investigation of Nonaqueous Phase Liquid Behavior at Heterogeneous Sand Layers Using VODA Multiphase Flow Code

Jiří Mikyška,1 Michal Beneš,1 and Tissa H. Illangasekare2

1Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University in Prague, Trojanova 13, 120 00 Prague, Czech Republic
2Center for Experimental Study of Subsurface Environmental Processes, Colorado School of Mines, 1500 Illinois St., Golden, CO 80401, USA

E-mail: jiri.mikyska@fjfi.cvut.cz

ABSTRACT

Interplay between the gravitational and capillary pressure forces at the interfaces of sands with different capillarity properties determines the fate of nonaqueous phase liquids (NAPLs) in the subsurface. Competition of these two forces can be observed on inclined interfaces of homogeneous sand formations. We have developed a multiphase flow code that has been applied to the study of NAPL behavior at the inclined interfaces. This model provides two methods for numerical treatment of sharp interfaces between the sands with different capillarity properties. Numerical results are presented indicating domains of applicability of these methods and their ability to describe discontinuous saturation profiles across the interface. The model is applied for computation of two-phase flow in a layered medium containing an inclined interface between two homogeneous layers with different capillary pressure parameters. As another application of the model, we present results of simulation of NAPL flow in a random medium consisting of small inclined homogeneous blocks of sands. Numerical results are compared to the results of laboratory experiments.

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1. INTRODUCTION

Two-phase flow models can be used for simulation of behavior of dense, nonaqueous phase liquids (NAPLs) in the saturated zone. As pointed out by several authors (Illangasekare et al., 1995; Helming, 1997; Kueper and Frind, 1991a, 1991b), medium heterogeneity has a profound effect on the fate of NAPLs in the subsurface, especially due to the entry pressure barrier effect that takes place at sharp soil interfaces separating subdomains with different capillary pressure–saturation curves. Due to this effect, the state variables of the two-phase flow system (phase pressures and phase saturations) can become discontinuous across the interface (de Neef and Molenaar, 1997). This has to be taken into account in the multiphase flow models so that they can reproduce the entry pressure effect correctly.

In order to incorporate the entry pressure effect into two-phase flow simulation codes, two methods can be used. The first one is based on the upwinding of the mobility coefficients (Forsyth, 1991; Helming, 1997), while the second one splits the computational domain into several homogeneous subdomains coupled together via some interface conditions, allowing for exact representation of the discontinuous saturation profiles at the interface. This idea was originally proposed (Bastian, 1999) within the framework of the finite volume box method.

We have developed an incompressible two-phase flow code in which we implement both methods within the framework of the control-volume finite-element scheme. We have shown previously (Beneš et al., 2006) how the latter method can be used within the framework of the control-volume finite element method. In this paper we show two examples of computations indicating that each method offers some advantages in different situations. Results of numerical simulations are compared to results of laboratory experiments.

2. MODEL EQUATIONS

Equations describing the multiphase flow in porous media can be developed from the Darcy laws and continuity equations for all involved phases (Helming, 1997). The presentation in this article is carried out for the system of two-phase equations in the following water pressure–NAPL saturation form:

$$\frac{\partial \phi}{\partial t} = \text{div} \left( \frac{k_{rw}(S_n)}{\mu_w} \rho_w K \cdot (\nabla p_w - \rho_w g) \right) + \rho_w q_w$$

$$\frac{\partial \phi}{\partial t} = \text{div} \left( \frac{k_{rn}(S_n)}{\mu_n} \rho_n K \cdot (\nabla p_n + \nabla p_c(S_n) - \rho_n g) \right) + \rho_n q_n$$

(1)

In these equations $\phi$ denotes the porosity, $\rho_w$ and $\rho_n$ are the phase densities, $S_n$ is the nonwetting phase saturation, $q_w$ and $q_n$ are the source/sink terms, $K$ stands for the intrinsic medium permeability tensor, $k_{rw}$ and $k_{rn}$ are the relative permeabilities of the respective phases, $\mu_w$ and $\mu_n$ are the viscosities of water and NAPL phase, and finally, $p_w$ and $p_c$ denote the water pressure and capillary pressure between the phases, respectively. Capillarity is modelled by the Brooks and Corey (1964) model, given by the following equation:

$$p_c(S_w) = p_d \cdot \left( \frac{S_w}{1 - S_w} \right)^{-1/\lambda}$$

where $S_w = \frac{S_w - S_{wr}}{1 - S_{wr} - S_{nr}}$ (2)

which is used in conjunction with the Burdine (1953) model for relative permeabilities. Here, $S_w = 1 - S_n$ is the wetting-phase saturation, $p_d$ is the entry pressure, $\lambda$ is the soil size distribution index, and $S_{nr}$ is the $\alpha$-phase irreducible saturation. The equations are to be solved in a bounded spatial domain $\Omega$ and in a time interval $(0, T)$. The initial and boundary conditions read as:

$$S_n(x, 0) = S_n^{ini}(x) \quad \forall x \in \Omega$$

$$p_w(x, t) = p_w^{Dir}(x, t) \quad \forall (x, t) \in \partial \Omega_{Dir} \times (0, T)$$

$$S_n(x, t) = S_n^{Dir}(x, t) \quad \forall (x, t) \in \partial \Omega_{Dir} \times (0, T)$$

(3)

$$v_n(x, t) = v_n^{Neu}(x, t) \quad \forall (x, t) \in \partial \Omega_{Neu} \times (0, T)$$

where $\partial \Omega_{Dir}$ and $\partial \Omega_{Neu}$ decompose the boundary
\[ \partial \Omega \] into a part with prescribed water-pressure/NAPL saturation (i.e., Dirichlet boundary condition, Dir) and a part with prescribed flux (i.e., Neumann boundary condition, Neu).

3. INTERFACE CONDITIONS

In heterogeneous media special conditions apply at the interfaces between layers of sands with different capillarity properties. As the capillary pressure–saturation curve can be position-dependent, i.e., \( p_c = p_c(S_w; x) \), we have to formulate conditions describing the two-phase flow behavior at the interfaces. These conditions were investigated in (de Neef and Molenaar, 1997). Assuming that there are no sources or sinks at the interfaces, the mass conservation law requires continuity of normal components of both phase fluxes \( \rho_w v_w \cdot \vec{n} \) and \( \rho_n v_n \cdot \vec{n} \) across the interface. Similarly, the wetting phase pressure should stay continuous at the interfaces. On the other hand, the continuity of the capillary pressure is possible only if there is a sufficiently high nonwetting phase saturation accumulated at the interface, so that its corresponding capillary pressure is greater or equal to the entry pressure of the soil on the opposite side of the interface. If not enough NAPL is accumulated at the interface, then the interface is impermeable (barrier effect). As the capillary pressure–saturation curves are different at both sides of the interface, the NAPL saturation is generally discontinuous across the interface (see Fig. 1). The nonwetting phase saturation at the higher entry pressure side (denoted as \( S_w^{II} \)) can be evaluated in terms of the saturation at the lowest entry pressure side of the interface (denoted as \( S_w^I \)) using the following condition

\[
S_w^{II} = \begin{cases} 
1 & \text{if } p_c^I(S_w^I) \leq p_d^I \\
\left(\frac{p_c^I}{I} - 1\right) & \text{if } p_c^I(S_w^I) > p_d^I 
\end{cases}
\]

4. NUMERICAL METHODS

Having covered the domain by a triangulation and denoting the linear basis functions corresponding

![Figure 1](attachment:image.png)

**Figure 1.** Capillary pressure–saturation curves of coarse sand (I), and fine sand (II). To pass through a heterogeneity interface from sand I to sand II, NAPL needs to reach the saturation \( S_w^{crit} \) in zone I at which the capillary pressure attains the entry pressure in zone II. Once NAPL passes the interface, the capillary pressure at the interface should be the same from both sides, which generally leads to different values of saturations \( S_w^I \) and \( S_w^{II} \) on both sides of the interface. If the capillary pressure in zone I is not high enough to attain the entry pressure in zone II, NAPL cannot penetrate into zone II.
to the triangulation by $N_i$, the following Control-Volume Finite-Element (CVFE) scheme (Forsyth, 1991; Helmig, 1997) can be derived as

$$
(1)^{h_{n_i}} \sum_{j \in n_i} \lambda_{ij} \rho_{ij} \gamma_{ij} (\psi^{k+1}_j - \psi^{k+1}_i) \\
+ \frac{\Delta t_k}{\rho_{ij} \gamma_{ij}} (\psi^{k+1}_j - \psi^{k+1}_i)
$$

(5)

where $n^{k+1}_{i,j} = \int_{\partial L_i} N_i \rho_{ij} \gamma_{ij} dS$ is the linear finite element discretization of the Neumann boundary condition term, $\psi^{k+1} = p^{k+1} + \delta_{i,n} p^{k+1} - p^{k+1}$ is the flow potential, and $\gamma_{ij} = -\int_{\partial L_i} \nabla N_i \cdot K \cdot \nabla N_j dS$ is the finite-element stiffness matrix. Taking values at the right-hand side at time $t_{k+1}$ leads to a fully implicit CVFE scheme. Furthermore, the mobility coefficients between nodes are chosen as the mobility in the upwind node, i.e., $\lambda_{i,j} = \lambda_{i,j}$ if $\gamma_{ij} (\psi_{ij} - \psi_{ji}) \leq 0$, and $\lambda_{i,j} = \lambda_{ji}$ in all other cases. Finally, $\Delta t_k$ denotes the $k$-th time step, and $V_i = \int_\Omega N_i dx$. The nonlinear discrete equations are linearized using the Newton-Raphson method with a line-search option. The resulting linear systems are solved using a linear multigrid solver as a preconditioner for the BiCGStab method (Van der Vorst, 1992; Saad 1998).

5. TREATMENT OF HETEROGENEITY

There are two methods of heterogeneity treatment that can be implemented in multiphase flow codes. The first of them is the upwinding of the mobility coefficients described above. As it was shown (Beneš et al., 2006), this method works well provided that each mesh node is assigned uniquely into a subdomain. This method has the advantage that the interface positions need not be explicitly described. The method simulates the capillary pressure barrier effect well, but a false linear interpolation of the saturation appears in the vicinity of the interfaces, where the saturation is expected to be discontinuous.

An alternative method can be used that accounts for the discontinuous saturation at the interfaces. This method can be used only if the interfaces are aligned with the mesh edges. At each interface node more values of saturation are allowed, depending on from which subdomain the node is actually approached. The appropriate value is then used for assembling of the Jacobian and the defect in the Newton method. As all saturation values at a node are coupled together via the interface conditions, it follows that only one value of saturation per node, namely, the saturation in the region with the lowest entry pressure, has to be stored in the memory and used as a primary variable in the Newton method, while the other saturation values can be evaluated using the interface conditions each time when needed. This method has the advantage of representing exactly the discontinuous saturation profile across the interface. The utility of both approaches is demonstrated in the following experiments.

6. VODA MODEL DEVELOPMENT AND TESTING

The CVFE scheme, together with both the abovementioned methods for treatment of heterogeneous interfaces, has been implemented into a new multiphase flow code VODA. VODA is written in C language using the UG library (Bastian et al., 1997).

This code was first tested on simplified 1D problems with known analytical solutions (McWhorter and Sunada, 1992; Chen et al., 1992; and McWhorter and Sunada, 1992) to verify the convergence of numerical results toward the exact solution. Results of the convergence analysis are available (Mikyška et al., 2004). For this study, generalized benchmark problems were used which have been described previously (Beneš et al. 2004, Fučík et al. 2005, and Fučík et al. 2007).

Both methods treating the heterogeneities have been described and compared to each other in detail (Beneš et al., 2006). The sensitivity of the model results with respect to the perturbations of capillary pressure parameters has been studied recently (Mikyška and Illanagasekare, 2007).
7. APPLICATION 1: NAPL FLOW AROUND AN INCLINED INTERFACE

We describe two examples of simulations for which there are laboratory data available. In the first computation we simulate incompressible two-phase flow in a rectangular domain consisting of two homogeneous layers separated by a straight inclined interface (see Fig. 2). This corresponds to a laboratory experiment described previously (Fagerlund et al., 2007a). The domain is fully saturated by water, and a slow flow from right to left is enforced by different heads imposed on the left and the right boundaries. The top and bottom parts of the boundary are permeable. There is a source of NAPL in the bottom layer that is released continuously at a constant rate. As the NAPL density is lower than water, NAPL is driven by the buoyancy force toward the interface. The entry pressure in the top layer is higher than in the bottom one, and thus NAPL accumulates below the interface. The question is whether a sufficient saturation of NAPL accumulates at the interface so that it can penetrate into the top layer. The values of all parameters are given in Tables 1 and 2. As the geometry of the domain is quite simple, the sharp-interface approach can be used. A coarse mesh can be prepared such that the interface is aligned with mesh edges. The computational mesh is obtained by taking two refinements of the coarse mesh (each refinement means that every triangle is subdivided into four smaller triangles by connecting the midpoints of edges). The resulting NAPL saturation distribution 3.5 hours after the NAPL injection started and the results of the corresponding laboratory experiment are shown in Figs. 3 and 4.

8. APPLICATION 2: NAPL FLOW IN STOCHASTICALLY GENERATED MEDIUM

In the second computation we simulate incompressible two-phase flow in a rectangular domain through a heterogeneous layer that is inclined and located be-

![Image of domain and boundary conditions](image-url)

**Figure 2.** Domain and boundary conditions for application 1 with two different homogeneous layers separated by a sharp inclined interface. Sand #30 is coarser (more permeable) than the sand #70. For sand parameters, see Table 2.

<table>
<thead>
<tr>
<th>Fluid</th>
<th>Density $\rho_\alpha$ (kg m$^{-3}$)</th>
<th>Viscosity $\mu_\alpha$ (kg m$^{-1}$ s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>W</td>
<td>999.7</td>
<td>0.0010</td>
</tr>
<tr>
<td>N</td>
<td>830.0</td>
<td>0.0035</td>
</tr>
</tbody>
</table>
Table 2
Sand parameters used for simulation. All values except those of $S_{nr}$ were measured in the laboratory experiment (Fagerlund et al., 2007b). The values of $S_{nr}$ are estimated (Fagerlund, 2004) by NAPL saturations observed after a long time in other previous experiments with the same sands and liquids.

<table>
<thead>
<tr>
<th>Sand</th>
<th>$K$ (m$^2$)</th>
<th>$\phi$</th>
<th>$p_d$ (Pa)</th>
<th>$\lambda$</th>
<th>$S_{nr}$</th>
<th>$S_{ur}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>8</td>
<td>$1.7 \times 10^{-9}$</td>
<td>0.43</td>
<td>363</td>
<td>4.8</td>
<td>0.11</td>
<td>0.30</td>
</tr>
<tr>
<td>16</td>
<td>$6.1 \times 10^{-10}$</td>
<td>0.40</td>
<td>484</td>
<td>2.77</td>
<td>0.06</td>
<td>0.30</td>
</tr>
<tr>
<td>30</td>
<td>$1.8 \times 10^{-10}$</td>
<td>0.43</td>
<td>1051</td>
<td>3.28</td>
<td>0.06</td>
<td>0.25</td>
</tr>
<tr>
<td>50</td>
<td>$6.6 \times 10^{-11}$</td>
<td>0.40</td>
<td>1380</td>
<td>3.04</td>
<td>0.11</td>
<td>0.15</td>
</tr>
<tr>
<td>70</td>
<td>$2.2 \times 10^{-11}$</td>
<td>0.44</td>
<td>2419</td>
<td>2.70</td>
<td>0.06</td>
<td>0.15</td>
</tr>
</tbody>
</table>

Figure 3. NAPL saturation distribution after 3.5 h of continuous injection as computed by the numerical model with the sharp interface approach.

Figure 4. Resulting NAPL plume as revealed in a laboratory experiment. The photograph is taken from Fagerlund et al. (2007a)

tween two homogeneous ones. The heterogeneous layer consists of small $3 \times 1$ cm bricks of five different sands. The exact distribution of the sands together with boundary conditions is shown in Fig. 5. Fluids and sands parameters that were used for the computations are shown in Tables 1 and 2. They approximately correspond to the values measured in a laboratory experiment. The detailed description of measurement and more precise parameter values are available (Fagerlund et al., 2007b).

At the beginning, the domain is fully saturated by water and a slow flow from right to left is enforced by the imposed heads on the left and the right boundaries. The top and bottom boundaries are again impermeable. At time $t = 0$, NAPL starts to be injected into the domain continuously up to $t = 9490$ s at a constant flux and then the influx is stopped. The final time of the simulation is 3.5 h (12600 s). Being lighter than water, NAPL is driven up through the heterogeneous layer, but in this case, its path is determined mainly by the complex heterogeneity field.

As the heterogeneity architecture is quite complex, we use the upwinding method of treatment of the heterogeneity so that there is no need to construct mesh whose edges would respect all the heterogeneity interfaces. The coarse mesh is shown in Fig. 6. The computational mesh is obtained by two refinements of the coarse mesh. This leads to a deformation of the real heterogeneity field caused by a projection on the mesh. Therefore, the computation uses the projected heterogeneity field that is shown in Fig. 7.

The computation turned out to be more complicated than in the previous example. Not only did the numerical algorithm require more time and memory to
Figure 5. Sand distribution and boundary conditions for the NAPL flow in random medium

Figure 6. A coarse mesh used for the NAPL flow in random medium. The actual mesh is obtained by taking two regular refinements of this mesh

Figure 7. Resulting heterogeneity field obtained by the projection of the field from Fig. 5 on the twice refined mesh

process, but also the Newton’s method encountered serious difficulties after achieving time $t = 3448$ s. Due to the increasing number of unsuccessful attempts of the nonlinear solver to improve the nonlinear defect, an adaptive time-stepping procedure was eventually forced to reduce the time step size below the minimal allowed time $\Delta t_{\text{min}} = 10^{-3}$ s, leading to the failure of the numerical algorithm. Exploration of the last available result revealed occurrence of high NAPL saturations that were very close to the maximal saturations allowed by the residual water saturations. This is exactly the situation in which the capillary pressure–saturation
curve has the unbounded slope. Therefore, we used a regularized version of the Brooks–Corey relationship in which the capillary pressure curve at a close vicinity of the asymptote is replaced by a straight tangent line of high but finite slope. The resulting NAPL saturation distribution obtained on two-level refinement of the coarse grid using the regularized capillary pressure–saturation curve and the result of a laboratory experiment are shown in Figs. 8 and 9. The model results show remarkably good correspondence to the laboratory results. It was not necessary to adjust any model parameters to achieve this correspondence.

**Figure 8.** NAPL saturation distribution after 3.5 h as computed by the numerical model using the upwinding of the mobility coefficients

**Figure 9.** Resulting NAPL plume as revealed in laboratory experiment. The photograph is taken over from Fagerlund et al. (2007b)

### 9. CONCLUSIONS

We have presented computational results of two numerical simulations obtained using VODA multiphase flow code and compared them to laboratory experimental results and achieved good agreement. The results also confirm utility and the correct function of both approaches used for treatment of heterogeneity interfaces in our numerical model.

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### REFERENCES


Bastian, P., Birken, K., Lang, S., Johannsen, K., Neuß, N., Rentz-Reichert, H., and Wieners, C., UG:
Investigation of NAPL Behavior in Heterogeneous Media using VODA


