

DNAPL SOURCE ZONE CHARACTERIZATION USING A STOCHASTIC AGGREGATION MODEL

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ABSTRACT

DNAPL source zone characterization is important for risk assessments, feasibility studies, and identification of appropriate remediation technologies. If sufficient DNAPL is released into the subsurface, it can penetrate the water table because of its relative density to water. When the DNAPL is also less viscous than water, an inherently unstable DNAPL-water displacement occurs. Descriptions of DNAPL source zones that accurately characterize these unstable pathways at the pore-scale have as yet been unattainable with commonly used modeling techniques.

Stochastic aggregation models are a variant of diffusion-limited aggregation models. These models have been used to model such unstable processes as dielectric breakdown, diffusion-controlled polymerization, chemical dissolution processes, solute leaching, fluid-fluid displacement in Hele Shaw cells, and viscous fingering in the absence of gravity in porous media. The authors of this paper have developed a stochastic simulation model for DNAPL source zone characterization. The proposed approach uses fluid properties, porous media properties, and macroscopic flow rates as inputs. This paper summarizes the approach used and compares model simulations to two-dimensional laboratory flume experiments.

Key words: DNAPLs, site characterization, modeling, groundwater

INTRODUCTION

Dense non-aqueous phase liquid (DNAPL) source zone characterization is important for risk assessments, feasibility studies, and identification of appropriate remediation technologies at DNAPL-contaminated sites. The extent and configuration of the source zone are also important inputs to multiphase mass transfer models. When a DNAPL displaces water, the interface between fluids is often unstable due to fluid and porous media properties. Solutions stemming from the continuity equation do not consider the stability of the interface at the pore scale. Using continuum models to determine fluid-fluid displacements at the pore scale is computationally prohibitive and beyond the capability of available numerical simulators (Poulsen and Kueper, 1992). However, many relevant questions need to be answered by pore-scale models. How much lateral spreading of the DNAPL can be expected? How much of the pore space was affected by the DNAPL? How much DNAPL is in contact with the groundwater? The model discussed in this paper can answer these questions.

The stochastic aggregation model (Trantham and Durnford, 1998) uses a modified DLA algorithm (Witten and Sander, 1983) to model the displacement of water by DNAPLs. Using the model, it is possible to look at the displacement of these fluids under different flow rates, changing porous media properties, and in a gravity field. The front configurations produced under these conditions range from viscous fingers to flat fronts. The model uses macroscopic, dimensionless capillary, and bond numbers to circumvent the difficulty of defining parameters on the microscale.

MODEL FORMULATION AND COMPARISONS

Instabilities in the fluid-fluid displacement are likely to arise on the pore scale when a non-wetting fluid invades a wetting fluid in a homogeneous porous medium (Homsy, 1987). In immiscible fluid flow, the capillary pressure, P_c , is defined as the difference between the pressure in the NAPL-phase and the pressure in the water:

$$P_c \equiv P_{nw} - P_w \quad (1)$$

Darcy's law can be written for each phase of a two-phase immiscible system as follows:

$$q_w = \frac{-kk_{rw}}{\mu_w} (\nabla P_w - \rho_w g \nabla z) \quad (2)$$

$$q_{nw} = \frac{-kk_{rnw}}{\mu_{nw}} (\nabla P_{nw} - \rho_{nw} g \nabla z) \quad (3)$$

where w denotes water; nw denotes NAPL; q is the volumetric flux (assumed positive down); μ is the dynamic viscosity; ρ is the density; P is the pressure; k is the intrinsic permeability; and kr is the relative permeability for each phase.

The capillary pressure gradient is solved for by combining Equations (1), (2), and (3):

$$\nabla P_c = \frac{1}{k} \left(\frac{q_w \mu_w}{k_{rw}} - \frac{q_{nw} \mu_{nw}}{k_{rnw}} \right) + \Delta \rho g \sin \alpha \quad (4)$$

where $\Delta \rho = \rho_{nw} - \rho_w$, and α is the angle of the flow from the horizontal. The product of k and k_{rw} is k_w and likewise for the NAPL phase. Equation (4) is analogous to the analysis of Saffman and Taylor (1958) and Chuoke et al. (1959).

The length scale and capillary pressure in Equation (4) are non-dimensionalized with macroscopic parameters following Hilfer and Aeren (1996). A hat designates the non-dimensional variables:

$$\hat{\nabla} = \nabla l \quad (5)$$

$$\hat{P} = \frac{P}{P_d} \quad (6)$$

Non-dimensionalizing the capillary pressure gradient results in Equation (7):

$$\hat{\nabla} \hat{P}_c = \frac{q_w \mu_w l a}{k_w P_d} - \frac{q_{nw} \mu_{nw} l a}{k_{nw} P_d} + \frac{\Delta \rho g l \sin \alpha}{P_d} \quad (7)$$

where a accounts for the change from pore scale to the macroscale. The stochastic aggregation algorithm is a pore-scale model. For the movement of the interface as described by Equation (4) to apply to the pore scale, a scaling factor of 10^2 must be included with the viscous terms (Morrow and Songkran, 1981; Baudet, et al., 1986; Dawson and Roberts, 1997).

The first two terms in Equation (7) are the macroscopic capillary number for water and NAPL, respectively. The third term is the macroscopic bond number as defined by Hilfer and Aeren (1996). Equation (7) is rewritten in terms of these dimensionless numbers:

$$\hat{\nabla} \hat{P}_c = \overline{Ca}_w - \overline{Ca}_{nw} + \overline{Bo} \quad (8)$$

where Ca_w and Ca_{nw} are the capillary numbers for water and NAPL, respectively, and Bo is the bond number. The bars denote that the dimensionless numbers are macroscopic. Equation (8) quantifies the competition between the capillary, viscous, and gravity forces at the interface. The scaled capillary pressure gradient in Equation (8) is termed the transition number, T , and is grouped as follows:

$$T = (\overline{Ca}_w - \overline{Ca}_{nw}) + \overline{Bo} \quad (9)$$

The first term of the transition number compares the macroscopic capillary numbers for the wetting and non-wetting phases and captures the combined effect of the viscous and capillary forces. The first term of the equation is positive for NAPLs that are less viscous than water and negative for NAPLs that are more viscous than water. The second term in the transition number is the macroscopic bond number and includes the effects of gravity forces due to the density difference between the NAPL and water. The second term is positive for DNAPLs and negative for LNAPLs, if the NAPL is displacing water from the top. Morrow and Songkran (1981) and Dawson and Roberts (1997) used linear combinations of the microscopic capillary and bond numbers for quantifying LNAPL and DNAPL entrapment, respectively.

The stochastic aggregation model follows a simple algorithm. When the simulation starts, particles are released from a random location opposite the seed point (the location where the DNAPL is injected). The particle has an equal probability of moving up, down, right, or left. The particle continues to walk randomly until it reaches a location adjacent to a location already occupied by a particle. The particle is incorporated into the aggregate with a probability equal to the sticking probability, and a new particle is released. Different sticking probabilities change the overall density of the aggregate. In the stochastic aggregation model, the sticking probability (Ps), accounts for the changes in front stability. A unique relationship between the sticking probability and the transition number was developed from 43 two-dimensional, DNAPL-water laboratory experiments. The relationship between the sticking probability and the transition number was determined as

$$Ps = 0.1227822|T|^{-1.3240317} \quad (10)$$

The model is validated for homogeneous porous media through comparison with laboratory experiments using Tetrachloroethylene (PERC), 1,2 Dichloroethane (DCA), Carbon Tetrachloride

(CTET) and Mobile Pyrogard 53 (made by Akzo Nobel, Gallipolis Ferry, WV, under the name Fyrquel 220). Examples are shown in Figure 1. The first three fluids are commonly used DNAPLs which are less viscous than water. Mobile Pyrogard 53 is a fire-resistant hydraulic fluid and is more viscous than water.

The laboratory experiments are shown on the left in Figure 1 with the corresponding model simulation shown on the right. It is important to compare the characteristics of the DNAPL-water displacement, i.e., branching, spreading, and relative thickness of the fingers.

The model is validated for heterogeneous porous media through comparison with the lab experiment of Kueper and Frind (1991). Figure 2 shows the heterogeneous sand packing used by Kueper and Frind (1991). PERC was allowed to infiltrate from the source area at the top of the flume under a constant head of 4 cm. Figure 3 shows the results of Kueper and Frind's (1991) lab experiment on the right with the corresponding model simulation on the left. The model simulations capture the characteristic DNAPL-water displacement and show that the PERC preferentially travels through regions of higher permeability.

CONCLUSIONS

The interface between DNAPL and water is often unstable due to DNAPL fluid properties relative to water. A stochastic aggregation model which simulates the movement of the DNAPL-water front on the pore scale was developed to better characterize DNAPL source zones. The model is currently well formulated for simulating two-dimensional DNAPL-water displacement for a variety of DNAPLs under the conditions of different inflow rates, different angles of inclination from the horizontal, and varying heterogeneous media properties. The comparison of model simulations to laboratory experiments remains limited to visual inspection, as is the case for other models currently attempting to model DNAPL-water displacements. Future work includes expanding the model to three dimensions.

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Figure 1. Application of model to DNAPL-water displacement in a two-dimensional flume. (a) PERC 90°, $Q=3 \text{ ml min}^{-1}$, $t=1 \text{ min}$, $T=0.354$, $Ps=0.485$. (b) 1,2 Dichloroethane 45°, $Q=6 \text{ ml min}^{-1}$, $t=1 \text{ min}$, $T=0.473$, $Ps=0.331$. (c) Carbon Tetrachloride 30°, $Q=6 \text{ ml}$

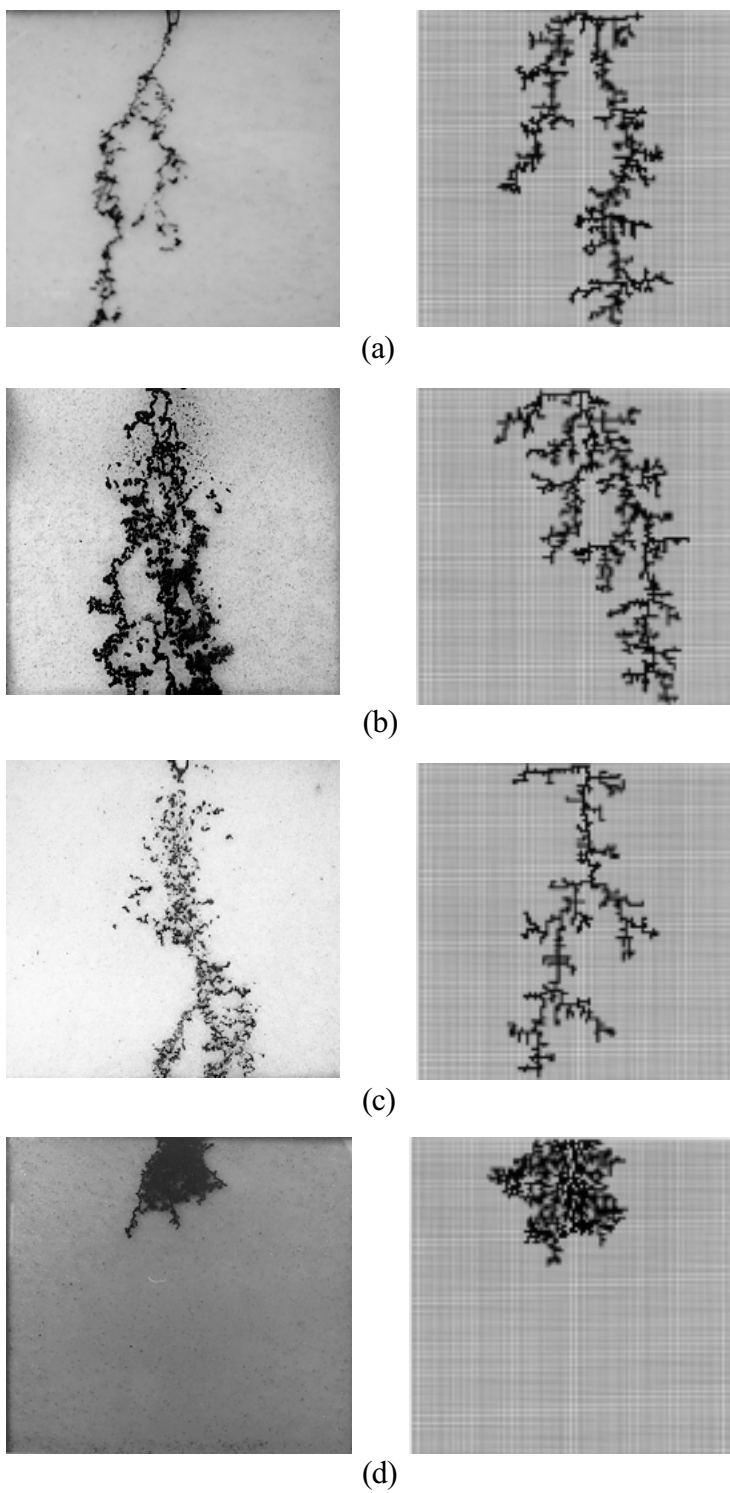


Figure 2. The heterogeneous sand packing of Kueper and Frind's (1991) PERC-water displacement experiment.

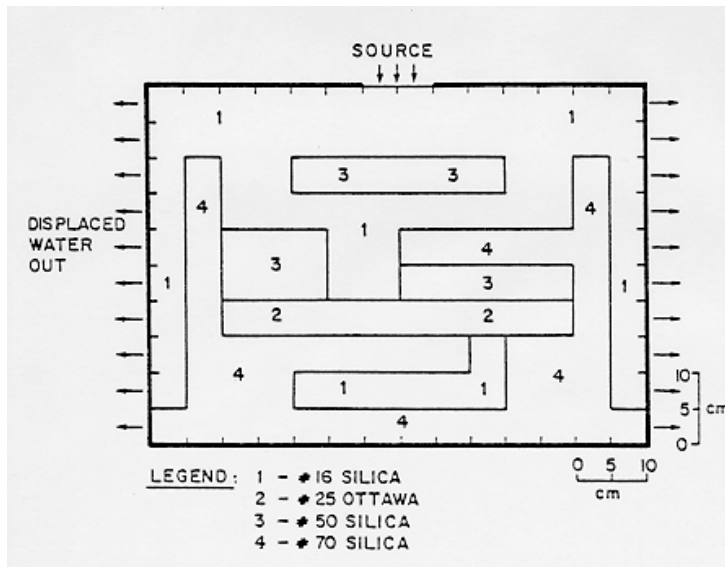


Figure 3. Model simulations (on the left) compared to the experimental results of Kueper and Frind (1991) shown on the right. (a) 34 seconds. (b) 126 seconds. (c) 245 seconds.

