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The influence of interfacial tension reduction on organic liquid migration: numerical and experimental comparisons

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Abstract The ability to simulate multiphase flow under conditions of surfactant influenced interfacial tension reduction was studied through comparisons with observed tetrachloroethylene (PCE) migration pathways in a heterogeneous laboratory sand tank system. In the first experiment, PCE migration in the absence of surfactant was observed to be impeded by less permeable lenses. Surfactants were used in a second experiment to produce very low interfacial tensions, resulting in PCE migration paths into and through the less permeable materials. A multiphase flow simulator incorporating traditional parametric models was successfully applied to predict the general PCE migration behaviour observed in the sand tank experiments. Migration pathways and final mass distribution, however, were found to be very sensitive to estimated fluid and soil properties, the capillary retention function selected, and capillary pressure scaling factors.

INTRODUCTION

Surfactant enhanced aquifer remediation (SEAR) is a promising innovative remediation technology for aquifers contaminated with dense, sparingly soluble, organic liquids (Pennell & Abriola, 1998). The use of surfactants to lower interfacial tension and enhance organic liquid mobility has been demonstrated to be highly effective in recovering organic liquids in laboratory soil columns. However, in field settings organic mobilization could create uncontrolled contaminant plunging or allow migration into less permeable materials. While numerical simulators offer a means to predict organic liquid mobility in heterogeneous systems and to assist in the design of SEAR applications, such models have yet to be verified for these applications.

The objective of this work is to examine the factors that affect the prediction of numerical multiphase flow under conditions of surfactant facilitated interfacial tension reduction. The accuracy of numerical multiphase flow simulations is assessed through the comparison of model predictions with observations from a series of infiltration experiments in laboratory sand tanks containing heterogeneous media.

LABORATORY EXPERIMENTS

Experiments of tetrachloroethylene (PCE) infiltration in laboratory sand tanks were conducted at the Georgia Institute of Technology (Singletary, 1998). The purpose of these experiments was to study the influence of interfacial tension reduction by surfactants on PCE infiltration pathways and entrapment. Figure 1 shows the dimensions and packing configuration of the sand tank.

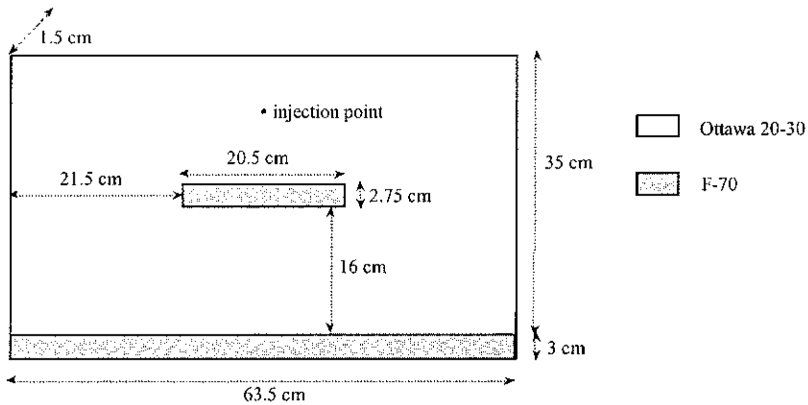

Fig. 1 Sand tank configuration.

The tank was packed with a 20-30 mesh Ottawa sand and a finer F-70 Ottawa sand. The F-70 sand was placed along the bottom of the tank and as a rectangular shaped lens positioned along the primary drainage pathway below the injection point. Hydraulic properties of the media are listed in Table 1.

Table 1 Soil properties used in simulations.

	Ottawa 20-30	F-70
Porosity	0.370 (a)	0.340 (a)
Permeability (m^2)	1×10^{-10} (a)	1×10^{-12} (a)
Residual water saturation	0.070 (b)	0.189 (c)
Residual organic saturation	0.176 (b)	0.206 (d)
Brooks-Corey air–water entry pressure (Pa)	654 (b)	4887 (c)
Brooks-Corey air–water pore size index	3.51 (b)	3.3 (c)
Air–water van Genuchten α (1/Pa)	0.00144 (b)	0.000241 (c)
Air–water van Genuchten n	7.42 (b)	4.3 (c)

(a) Singletary (1998); (b) Powers (1992); (c) Kueper & Frind (1991); (d) estimated.

The sand tank aquifer system was initially saturated with water or an aqueous-surfactant solution designed to yield a specific PCE/aqueous interfacial tension. The surfactant used in the work described here was a 4% solution of sodium dihexyl sulphosuccinate (Aerosol MA80i) and sodium dioctyl sulphosuccinate (Aerosol OT100) mixed in a 4:1 ratio (4% MA/OT), respectively (Singletary, 1988). This surfactant produced a nearly two order reduction in interfacial tension, which was measured at the Georgia Institute of Technology (Loverde, 1997). A known quantity of PCE was injected above the fine sand lens at a constant rate. The PCE migration pathway was visually recorded with time. After a final immobile PCE distribution was established, the tank was opened and destructive sampling was performed to quantify the PCE distribution. Table 2 lists fluid properties and conditions for the two experiments presented here.

Table 2 Properties and conditions of experiments.

	Experiment 1	Experiment 2
Pore fluid	water	4% MA/OT
PCE/aqueous interfacial tension (dyne cm^{-1})	47.8	0.5
PCE injection rate (ml min^{-1})	0.50	0.05
PCE injection volume (ml)	19.46	19.5

NUMERICAL SIMULATION

The two PCE infiltration experiments were simulated with a multiphase immiscible simulator, M-VALOR, an implicit pressure, explicit saturation (IMPES) based block-centred finite difference model (Abriola *et al.*, 1992). This code has been verified against analytical solutions, taking full account of capillarity (Demond *et al.*, 1996).

The simulation of immiscible flow requires the specification of capillary pressure-saturation and relative permeability-saturation functions. Two commonly used capillary functions were employed in this work to explore the influence of the retention function on solution behaviour. The Brooks-Corey (BC) model (Brooks & Corey, 1964) has a distinct entry pressure below which capillary pressure variation with saturation is ignored:

$$P_c = P_d \overline{S_w}^{(-1/\lambda)} \; ; \; P_c \geq P_d \tag{1}$$

where P_c is the capillary pressure; P_d is the entry pressure; $\overline{S}_w$ is the effective water saturation; and λ is the pore size index. The van Genuchten (VG) model (van Genuchten, 1980) is characterized by a continuous representation of capillary pressure over the full range of saturation:

$$P_c = \frac{1}{\alpha}\left(\overline{S_w}^{-1/m} - 1\right)^{1/n} \tag{2}$$

where α, m and n are fitting variables. Identical relative permeability relations were used in all simulations. Relative permeability functions are estimated with the approach of Kaluarachchi & Parker (1992):

$$k_{rw} = \sqrt{\overline{S_w}}\left[1 - \left(1 - \overline{S_w}^{1/m}\right)^m\right]^2 \tag{3}$$

$$k_{ro} = \sqrt{1 - \overline{\overline{S_w}}}\left(1 - \overline{\overline{S_w}}^{1/m}\right)^{2m} \tag{4}$$

where k_{rw} and k_{ro} denote the water and organic relative permeability, respectively, and $\overline{\overline{S_w}}$ is the apparent water saturation that accounts for hysteretic entrapment. A complete description of the relative permeability relations and hysteresis model is presented in Rathfelder & Abriola (1998). Note that in the simulations presented here, redistribution and hysteretic entrapment is considered only for experiment 1, and only for a short period at the end of the simulation.

To estimate capillary retention properties for the aqueous-surfactant solution, a traditional Leverett scaling approach was used:

$$P_{c-\text{org/water}} = P_{c-\text{air/water}} \frac{\left[\gamma \cos\theta_R Z(\theta)\right]_{\text{org/water}}}{\left[\gamma \cos\theta_R Z(\theta)\right]_{\text{air/water}}} \tag{5}$$

where γ is the interfacial tension, θ_R is the contact angle corrected for roughness, and $Z(\theta)$ is a curvature correction factor (Demond *et al.*, 1994). Because no measurements were available for contact angle and roughness, their effect was initially ignored in the simulations. However, Demond & Roberts (1991) and Demond *et al.* (1994) found that the inclusion of these correction factors is increasingly significant with diminishing interfacial tension.

Grid resolution has also been shown to influence solution behaviour in the simulation of multiphase flow (Rathfelder & Abriola, 1998). To reduce smearing of saturation profiles, a relatively fine grid was employed. The sand box domain was discretized into approximately 1 cm square blocks, with finer vertical discretization employed within and immediately above the F-70 sand lenses (up to 0.5 cm). This resolution provides more that five cells spanning the entry pressure height of the Ottawa 20-30 sand in experiment 1 ($\approx$4.4 cm water), but less than one cell spanning the entry pressure height in experiment 2 ($\approx$0.05 cm water).

RESULTS

Figures 2 and 3 compare the visual observations of PCE migration in experiment 1 with simulated results using the VG and BC retention functions. Note that the visual observations reflect the PCE migration pathways as observed on one side of the tank, but do not indicate the volume of PCE present and may not fully represent internal migration pathways. The observed migration pathways in experiment 1 show that PCE does not enter the F-70 sand. The PCE pools on top of, and flows around, the upper lens, eventually pooling on top of the bottom lens. Due to slight irregularities in the packing of the F-70 lenses, there is preferential PCE flow towards the right side of the upper lenses. The simulation model does not account for these irregularities and therefore simulated flow distributions are symmetric around the upper lens.

The simulation results shown in Fig. 3 are consistent with observed behaviour. Solutions with the VG and BC retention functions both predict that PCE does not enter the F-70 lens, instead forming pools and flowing around the upper lens. Predicted PCE saturations above the F-70 lenses generally agree with measured values obtained by destruction sampling. Measured saturations ranged from 8 to 19% in 2 by 2 cm squares, with the greatest values immediately below the injection point and the vertical edges of the upper lens. The predicted saturation values, averaged over 2 cm heights above the lenses, qualitatively agree with the measured quantities in magnitude and location.

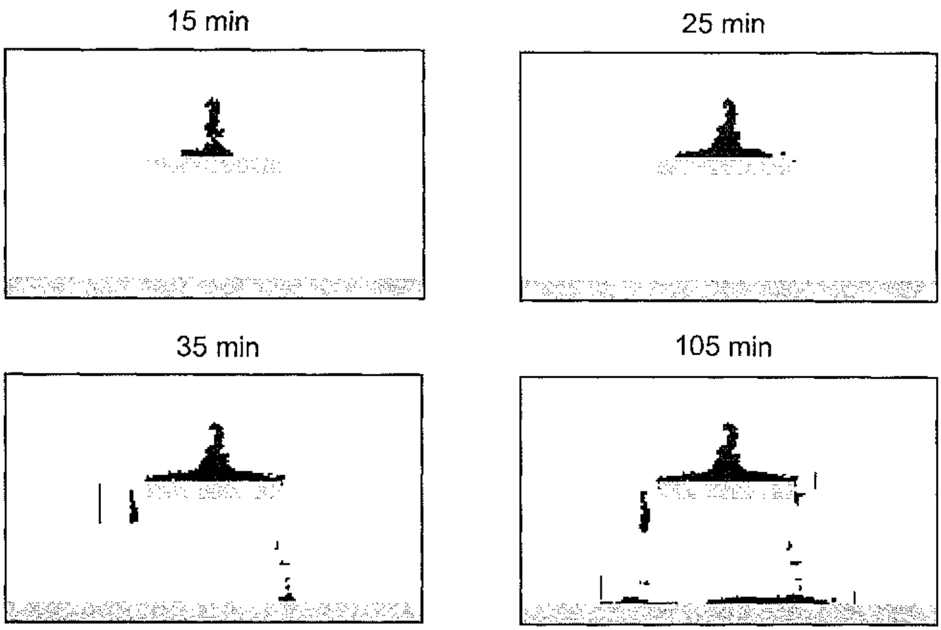

Fig. 2 Observed PCE distributions in experiment 1 (IFT = 47.8 dyne cm^{-1}).

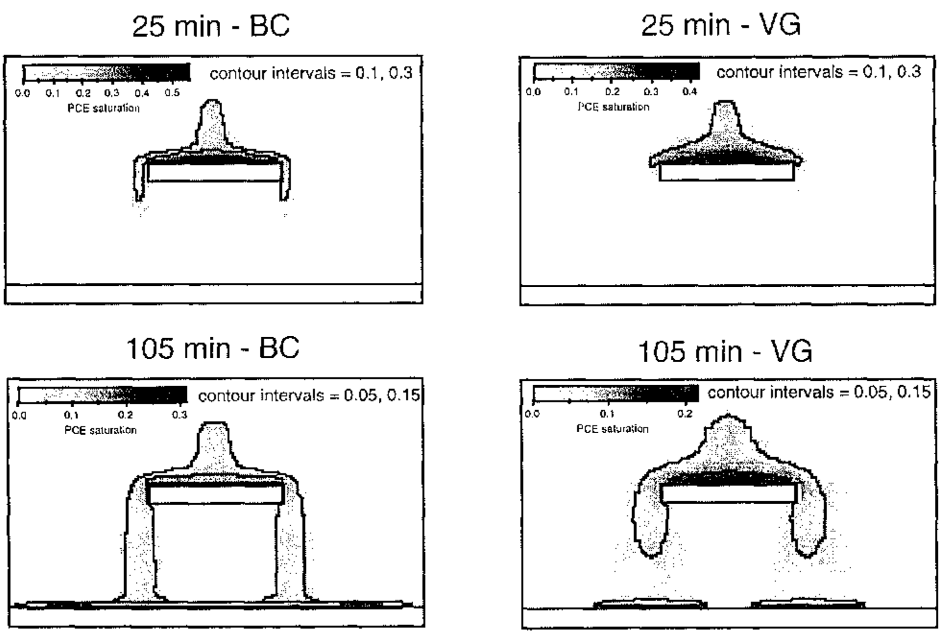

Fig. 3 Comparison of predicted PCE distributions in experiment 1 (IFT = 47.8 dyne cm^{-1}).

Closer examination, however, shows that there are differences in the simulation results obtained with the VG and BC retention functions. Consistent with findings of Rathfelder & Abriola (1998), numerical solutions with VG function exhibit greater

PCE spreading at low organic saturations. This is a consequence of the gradient of the retention function at low organic saturations, which is large in the VG function and small in the BC function. Due to these differences, the VG predictions exhibit a greater pooling height on the upper lenses and a slower overall migration rate. In contrast, results with the BC model show a more concentrated migration pathway, manifested by smaller pooling heights and a larger maximum saturation. Consequently the predicted overall migration rate is greater with the BC function and appears to outpace the observed rate.

Figures 4 and 5 compare the visual observations of PCE migration in experiment 2 with simulated results using the VG and BC retention functions respectively. In this experiment the very low interfacial tension of the MA/OT surfactant solution resulted in very little capillary resistance at the lens boundary. The PCE was observed to move easily and quickly through the Ottawa 20-30 mesh sand, and upon reaching the F-70 lens was rapidly able to overcome the entry pressure and flow through the fine grained soil. Flow through the upper lens appeared to occur in small fingers, which joined into three to four large channels upon exiting the lens. Flow through the F-70 lens was not completely unimpeded, however, with some of the PCE observed to pool on top of the upper lens and flow around on the right side. The measured height of the PCE pool on the upper lens was approximately 0.2–0.3 cm, which is less than the vertical grid spacing of 0.5 cm above the lens

Simulation results in Fig. 5 demonstrate that the traditional Leverett scaling approach (equation (5)) was able to sufficiently reduce the entry pressure to enable PCE migration through the F-70 lens. However, the simulation results reveal that the F-70 lens provided less impedance than was observed, exhibiting a small amount of pooling on top of the lens and no flow around the lens. Consequently the overall PCE migration rate in the simulated case is greater than the observed rate. Simulation results with the VG and BC retention functions exhibit only minor differences. This could be a consequence of insufficient grid resolution, which is greater than the entry pressure height. Overall the results are encouraging in that flow into the fine lens was predicted. Major differences between the observed and predicted distributions are the lack of pooling and flow around the upper lens in the simulated case, resulting in a greater overall migration rate, and the observed fingering behaviour through the lens which was not replicated by the model.

Errors in the traditional Leverett scaling approach (equation 5) are known to increase with diminishing interfacial tension (Demond & Roberts, 1991). Therefore, additional simulations were performed for experiment 2 to examine the sensitivity of solution behaviour to the scaling factor. Work by Demond & Roberts (1991) and Demond *et al.* (1994) indicates that there should be a positive correction to the scaling factor under drainage conditions. To account for roughness and contact angle variations the scaling factor was increased by a factor of 1.25 to 2, which corresponds to increases in the interfacial tension from 0.5 dyne cm^{-1} to a range of 0.625 to 1.0 dyne cm^{-1}.

Figure 6 reveals that small changes in the scaling factor can dramatically alter predicted PCE distributions in comparison to those shown in Fig. 5. In all cases, a small increase in the scaling factor enhanced pooling and spreading over the upper lens. At a certain level, PCE migrated both through and around the upper lens as was

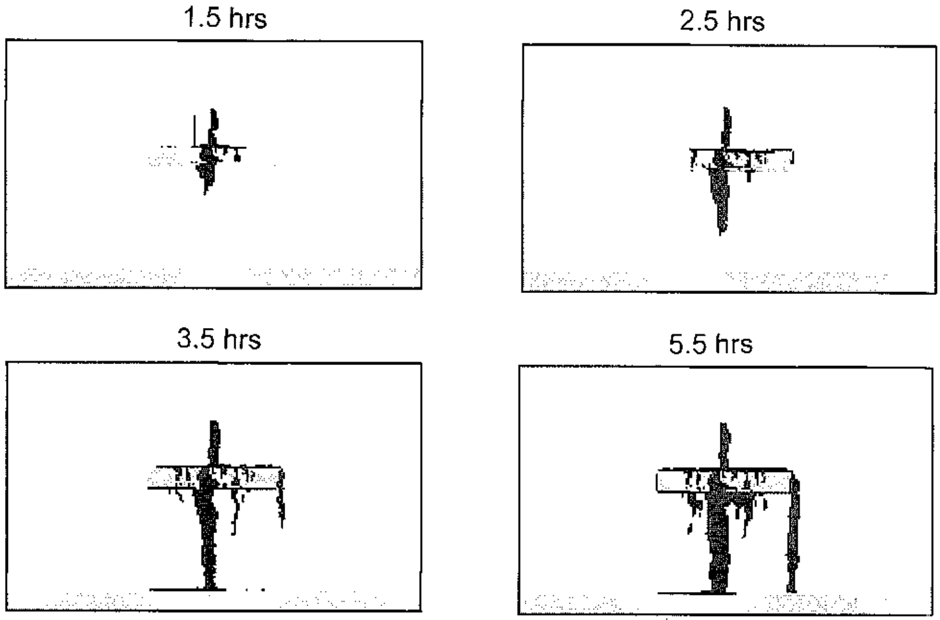

Fig. 4 Observed PCE distributions in experiment 2 (IFT = 0.5 dyne cm^{-1}).

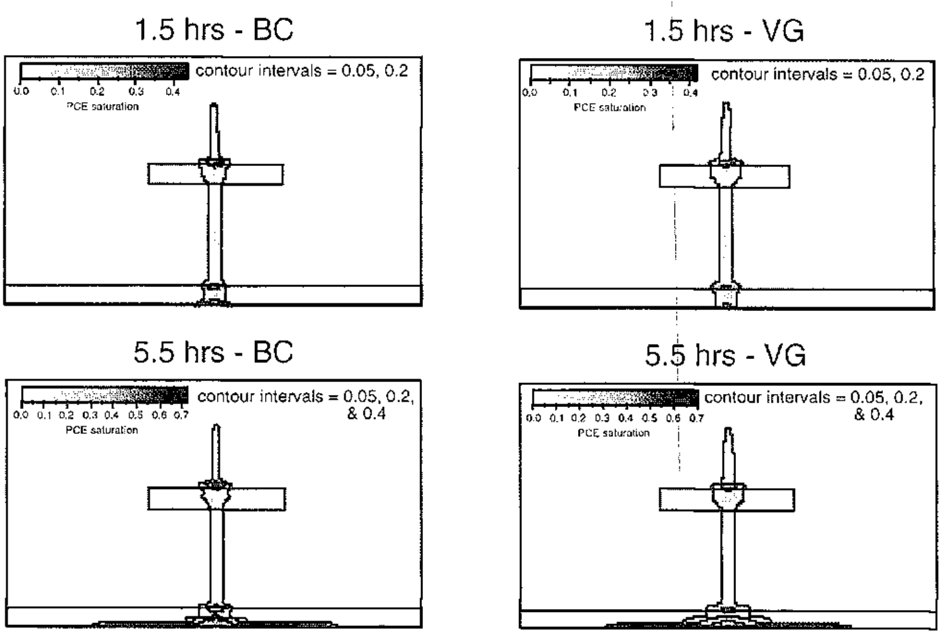

Fig. 5 Comparison of predicted PCE distributions in experiment 2 (IFT = 0.5 dyne cm^{-1}).

observed in the experiment. The change in the scaling factor necessary to obtain this behaviour was much smaller with the BC model (25%) than with the VG model (100%). This behaviour is consistent with results of Rathfelder & Abriola (1998) who showed that a numerical characteristic of the VG model is a greater propensity to enter finer grained material. In the extreme example, an increase in the scaling factor of 50% and use of the BC model completely alters the solution behaviour from almost no impedance by the F-70 lens to nearly complete bypassing around the F-70 lens. Note that the change in interfacial tension required to increase the scaling factor by 50% is very small (γ increased from 0.5 to 0.75 dyne cm^{-1}). Several factors could potentially contribute to small changes in the scaling factor, including required corrections as pointed out by Demond & Roberts (1991), errors in the measurement of interfacial tension, or experimental errors in conducting the sand box experiments.

Predictions in Fig. 6 also show that flow around the upper lens occurs at small PCE saturation. It is hypothesized that the lack of a concentrated flow path around the lens is a result of smearing of the saturation profile due to inadequate grid resolution. The horizontal grid spacing adjacent to the upper lens is 1 cm, which is much greater than the entry pressure height (≈0.05 cm water). Grid resolution was shown to significantly affect flow behaviour by Rathfelder & Abriola (1998). Observed PCE migration behaviour also indicated the presence of concentrated flow paths or fingers through the upper lens. These are likely the result of micro-heterogeneities along the sand interface, which cannot be characterized. Consequently fingering behaviour cannot be simulated with this model.

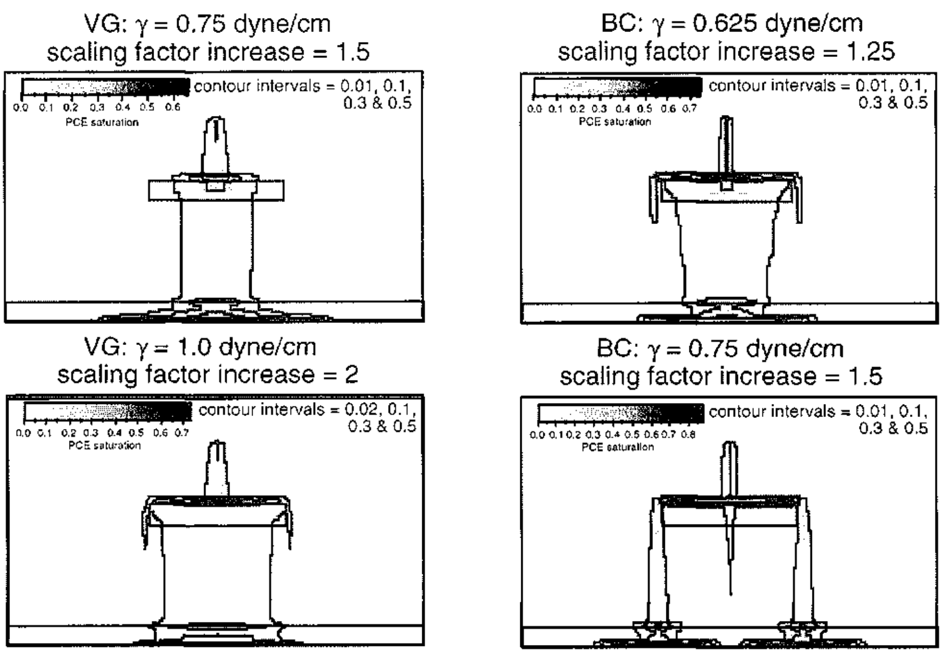

Fig. 6 Comparison of predicted PCE distributions in experiment 2 (IFT = 0.5 dyne cm^{-1}) at time = 5.5 hours using varying scaling factors.

CONCLUSIONS

Numerical models for immiscible fluid flow and existing parametric models for capillary retention and relative permeability can be successfully used to simulate the general migration behaviour of surfactant enhanced organic liquid migration.

However, the degree of accuracy depends on the ability to characterize fluid and soil properties. Solution behaviour also depends on numerical criteria including specification of the capillary retention function and grid resolution. At very low interfacial tensions, numerical solutions are highly sensitive to the capillary pressure scaling factor.

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