Effect of S-p Relation Model on DNAPL Migration Simulation Result

H. Ishimori *[1] and K. Endo [1]
[1] National Institute for Environmental Studies, Japan
*Corresponding author:
16-2, Onogawa, Tsukuba, Ibaraki, 305-8506, Japan, ishimori.hiroyuki@nies.go.jp

Abstract: To consider effective countermeasures against groundwater contaminated with dense non-aqueous phase liquids (DNAPLs) such as chlorinated solvents, it is first important to understand the mechanism of their migration in heterogeneous aquifer. In addition, numerical analysis models to simulate such a complex migration in heterogeneous aquifer are required. The displacement pressure, which is a minimum pressure for one liquid to enter a soil pore saturated with the other liquid, affects DNAPLs migration in heterogeneous aquifer. Although effects of the displacement pressure on DNAPLs migration are represented by the relationship between liquid saturation and capillary pressure (*S-p* relation) such as van Genuchten model or Brooks and Corey model, there are few reports on their compatibilities between real phenomena and simulation result. This study focused on the displacement pressure effects on the DNAPLs migration, and pointed out the difference between an experimental result and simulation results with each *S-p* model.

Keywords: Dense non-aqueous phase liquid, Displacement pressure, *S-p* model, Two-phase flow, Two-dimensional DNAPL migration test

1. Introduction

Dense non-aqueous phase liquids (DNAPLs) such as chlorinated solvents can easily permeate into aquifer, because they have a lower viscosity and higher fluid density than water. They have a potential to cause long-term and regional contamination in the groundwater environment. The groundwater contaminated with DNAPLs and their remediation have become a major environmental concern. In order to carry out effective countermeasures against groundwater contaminated with DNAPLs, it is first important to understand the mechanism of their migration in heterogeneous aquifer. In addition, numerical analysis models to simulate such a complex migration in heterogeneous aquifer are required, because it is necessary to beforehand evaluate the effectiveness of a countermeasure against the contamination.

In heterogeneous aquifer system, it is well known that the displacement pressure, which is a minimum pressure for one liquid to enter a soil pore saturated with the other liquid, significantly affects the simulated DNAPLs migration. The relation between liquid saturation and capillary pressure (*S-p* relation) such as van Genuchten model (VG model) or Brooks and Corey model (BC model) is important in DNAPLs migration simulation. The VG model is widely used in most previous studies and commercial softwares in DNAPLs migration simulations. But, there are few reports on the compatibilities of their model between actual phenomena and simulation result as to DNAPLs migration in heterogeneous aquifer system. Hence, it should be investigated how the difference between VG model and BC model has an influence on the simulation results, and their simulation results should be also compared with experimental results to evaluate the accuracy of the simulation model.

In this study, the simulations of DNAPLs migration in a heterogeneous saturated aquifer were conducted, and their results were compared with the experimental result previously reported by Kamon, M. et al. (2004) as shown in Figure 1.

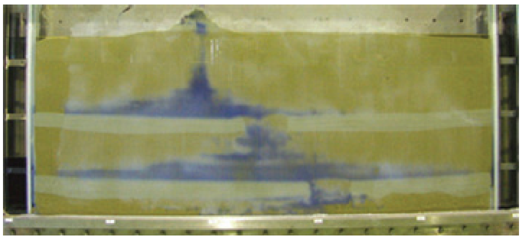


Figure 1. DNAPL migration experiment in heterogeneous aquifer. It is HFE distribution after 8 hours. The experimental conditions were as follows: The tank had a 0.65 m height and 1.50 m width. The aquifer consisted of coarse sand and fine sand. HFE-7100, which simulated a tetrachloroethylene, was continuously injected at a constant pressure of 5.4 kPa.

2. Theory and Governing equations

2.1 Governing equations

The governing equations for water and oil two-phase flow system are represented as

$$\phi \frac{\partial S_w}{\partial t} + \nabla \cdot \left[-\frac{k_{rw}\mathbf{K}}{\mu_w} \left(\nabla p_w + \rho_w g \nabla z \right) \right] = 0 \tag{1}$$

$$\phi \frac{\partial S_o}{\partial t} + \nabla \cdot \left[-\frac{k_{ro}\mathbf{K}}{\mu_o} \left(\nabla p_o + \rho_o g \nabla z \right) \right] = 0 \tag{2}$$

where, the subscripts, "w" or "o", of parameters show "water" or "oil", respectively. S_α is the degree of the saturation for the α fluid (α = w, o). ρ_α (kg/m^3) is the fluid density, μ_α (Pa s) is the viscosity, p_α (Pa) is the pressure, and $k_{r\alpha}$ is the relative permeability for the α fluid. ϕ is the porosity and $\mathbf{K}$ (m^2) is the intrinsic permeability. g (m/s^2) is the gravity acceleration and z (m) is the elevation vector. The governing equations shown in Eqs.(1)-(2) includes four primary variables of S_w, p_w, S_o, and p_o. Therefore, the numbers of their variables should be decreased to two by using two constraint conditions. One is a constraint on the fluid saturations.

$$S_w + S_o = 1 \tag{3}$$

And, the other is a constraint on the relationship between the fluid saturation and pressure.

$$S_w = f\left(p_{ow}\right) \tag{4}$$

where, p_{ow} (Pa) is the capillary pressure between the water phase and the oil phase. It is defined as $p_{ow} = p_o - p_w$. The *S*-*p* function of the capillary pressure, p_{ow}, to the water saturation, S_w, is determined from the water retention curve, and it is generalized by van Genuchten model (VG model) or Brooks and Corey model (BC model).

When the pressures of p_w and p_o are selected as the primary variables, the governing equations are rewritten using Eqs.(3)-(4) as follows:

$$-\phi \frac{\partial S_w}{\partial p_{ow}} \left(\frac{\partial p_w}{\partial t} - \frac{\partial p_o}{\partial t} \right) + \nabla \cdot \left[-\frac{k_{rw}\mathbf{K}}{\mu_w} \left(\nabla p_w + \rho_w g \nabla z \right) \right] = 0 \tag{5}$$

$$\phi \frac{\partial S_w}{\partial p_{ow}} \left(\frac{\partial p_w}{\partial t} - \frac{\partial p_o}{\partial t} \right) + \nabla \cdot \left[-\frac{k_{ro}\mathbf{K}}{\mu_o} \left(\nabla p_o + \rho_o g \nabla z \right) \right] = 0 \tag{6}$$

The governing equations should be solved for two fluid pressures of p_w and p_o under a certain initial and boundary conditions.

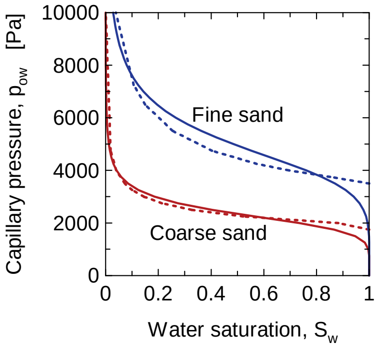


Figure 2. *S*-p models for coarse sand and fine sand. The solid lines show VG model and the dash lines show BC model, where α = 2.16 x 10^{-4} 1/Pa, β_{ow} = 2.044, n = 6.57, λ = 4.46, p_d = 3980 Pa for the coarse sand, and α = 1.04 x 10^{-4} 1/Pa, β_{ow} = 2.044, n = 5.70, λ = 3.16, p_d = 7280 Pa for the fine sand.

2.2 S-p relations

The *S*-*p* function shown in Eq.(4) is given by VG model (van Genuchten, 1980) or BC model (Brooks and Corey, 1964). When the residual saturation can be neglected, they are respectively

$$S_w = \left[1 + \left(\alpha \beta_{ow} p_{ow} \right)^n \right]^{-m} \tag{7}$$

$$S_w = \left(\frac{p_{ow}}{p_d} \right)^{-\lambda} \tag{8}$$

where, α (1/Pa), n, and m are the VG parameters. The m is determined as $m = 1-1/n$. p_d (Pa) is the displacement pressure, and it is approximately evaluated by $p_d \cong 1/(\alpha\beta_{ow})$. β_{ow} is the scaling factor, and it is given as $\beta_{ow} = \sigma_{aw}/\sigma_{ow}$. σ_{aw} (N/m) is the surface tension in air-water system, and σ_{ow} (N/m) is the interfacial tension in oil-water system. Figure 2 shows the *S*-p relations given by VG model or BC model. The VG model permits oil to enter the soil saturated with water even if its capillary pressure is so small. In contrast, the BC model permits oil to enter the soil only when its capillary pressure is more than the displacement pressure. These data in Figure 2 are for the coarse sand and fine sand.

2.3 k-S relations

The permeability depends on the presence of water and oil in the soil pore. The relative permeabilities, k_{rw} and k_{ro}, are also represented by VG model and BC model. When the residual saturation can be neglected, they are respectively

$$k_{rw} = S_w^{1/2}\left[1-\left(1-S_w^{1/m}\right)^m\right]^2 \tag{9}$$

$$k_{ro} = \left(1-S_w\right)^{1/2}\left(1-S_w^{1/m}\right)^{2m} \tag{10}$$

and

$$k_{rw} = S_w^{(2+3\lambda)/\lambda} \tag{11}$$

$$k_{ro} = \left(1-S_w\right)^2\left(1-S_w^{(2+\lambda)/\lambda}\right) \tag{12}$$

2.4 Analysis conditions

The two-phase flow analysis was conducted in two-dimensional domain, which had a 0.65 m height and 1.50 m width as shown in Figure 3. The domain consisted of the coarse sand and the fine sand. For the initial condition, the domain was fully saturated. For the boundary conditions, HFE which simulated a tetrachloroethylene was released at a constant capillary pressure of p_{ow} = 5.4 kPa from a point shown in Figure 3. In addition, the top and bottom boundaries were impermeable. The left boundary had a higher static water pressure than the right boundary, in order to simulate the groundwater flow with a certain hydraulic gradient. Table 1 summarized the analysis conditions. The physical properties were constant, and the groundwater velocity was parametrically changed. The purpose of this study is to investigate effects of the groundwater velocity and the *S*-*p* model function on the simulated distribution of the HFE, and to compare their simulation results with previous experimental result as shown in Figure 1.

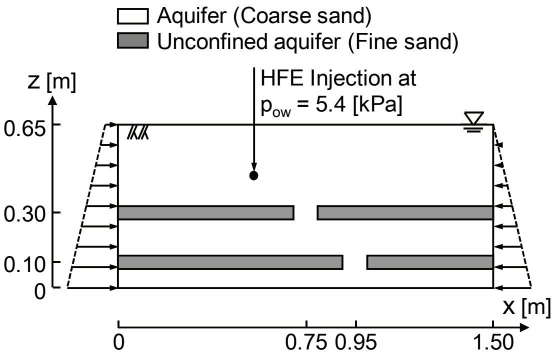


Figure 3. Two-dimensional analysis domain

Table 1. Analysis condition

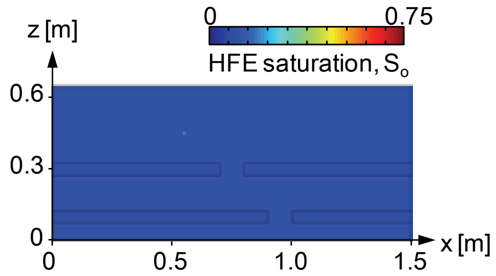
Parameter	Value		Unit
	Coarse sand	Fine sand	
ρ_w	1,000		kg/m^3
μ_w	1.00 x 10^{-3}		Pa s
ρ_o	1,520		kg/m^3
μ_o	1.00 x 10^{-3}		Pa s
β	2.04		
ϕ	0.30	0.30	
K	1.48 x 10^{-11}	2.37 x 10^{-12}	m^2
α	2.16 x 10^{-4}	1.04 x 10^{-4}	1/Pa
n	6.57	5.70	
λ	4.46	3.16	
p_d	3,980	7,280	Pa

2.5 Use of COMSOL Multiphysics

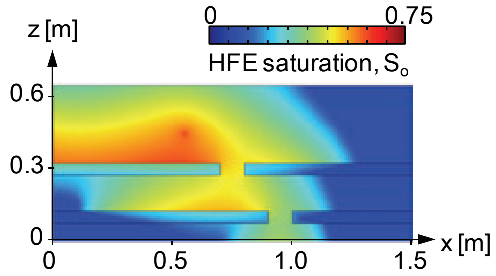
The simulations of the HFE migration were conducted using COMSOL Multiphysics ver 3.5a. Application mode was Darcy Law's pressure analysis in Earth Science Module. Two Darcy Law's Pressure Equations were used for the water-phase and the HFE-phase. In order to solve two-phase flow equations shown in Eqs.(5)-(6), the default equation system should be modified. The term of $\phi\,\partial S_w/\partial p_{ow}$ needed to be added to the Damping/Mass term in the default equation system. In this equation system with the primary variables of p_w and p_o, it may be difficult for COMSOL to converge a solution of an initial calculation stage that the HFE is injected to the aquifer fully saturated with the water. Therefore, an extremely little amount of the HFE was distributed in the initial analysis domain to improve the convergence.

3. Results and Considerations

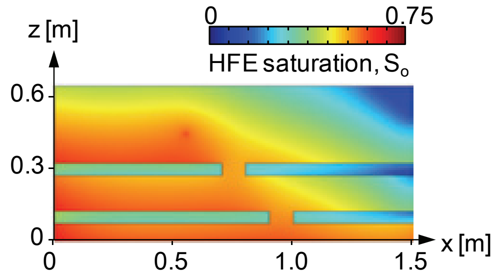
Figures 4 and 5 show the simulated HFE saturation distribution at 8 hours after the HFE was injected. Figure 4 was calculated from VG model, and Figure 5 was calculated from BC model. The remarkable difference in the HFE saturation distributions appeared. The HFE was distributed more widely in VG model than in BC model. The HFE infiltrated the unconfined aquifer in VG model, but did not infiltrate it at all in BC model. The simulation result by BC model was almost the same as the experimental result as shown in Figure 1. BC model does not permit the HFE to infiltrate a porous media in a condition that the capillary pressure is less than



(a) initial

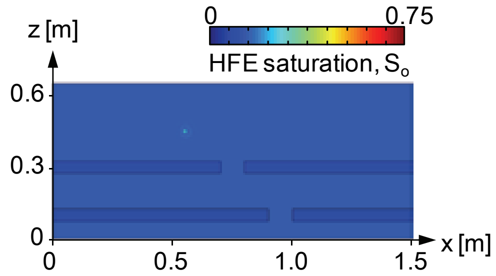


(b) after 4 hours

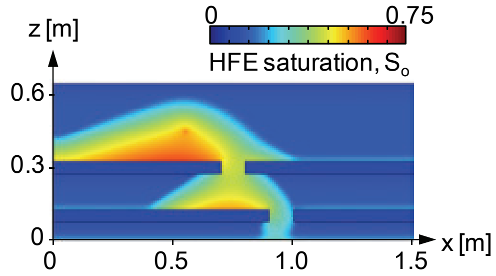


(c) after 8 hours

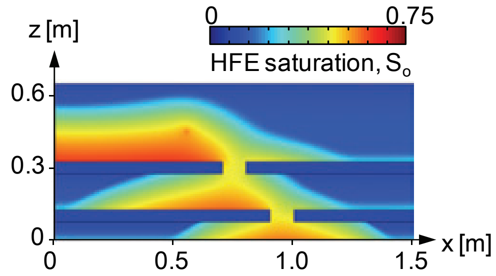
Figure 4. Simulated HFE saturation distribution using VG model without groundwater flow



(a) initial



(b) after 4 hours

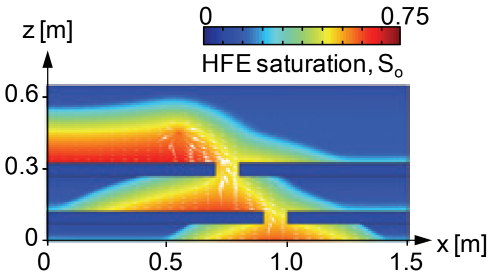


(c) after 8 hours

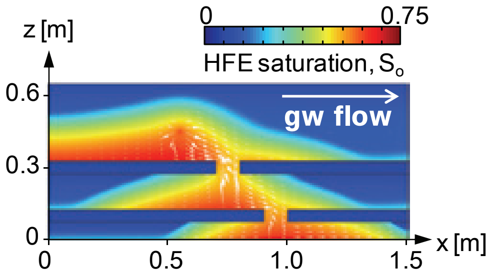
Figure 5. Simulated HFE saturation distribution using BC model without groundwater flow

the displacement pressure. In contrast, VG model permits the HFE to infiltrate a porous media regardless of the displacement pressure so that the HFE infiltrates an even porous media with a low permeability and high displacement pressure. Therefore, VG model simulated faster infiltration and wider distribution of the HFE than BC model under a condition of constant injection pressure. In case that the simulation is conducted under a condition of not constant injection pressure but constant injection rate, the remarkable difference like Figures 4 and 5 may not appear in the HFE saturation distribution. It is because the volume of the HFE which infiltrates the aquifer becomes the same regardless of the use of VG model or BC model. But, the difference of each model would appear in whether HFE can permeate unconfined aquifer with relatively low permeability and high displacement pressure or not.

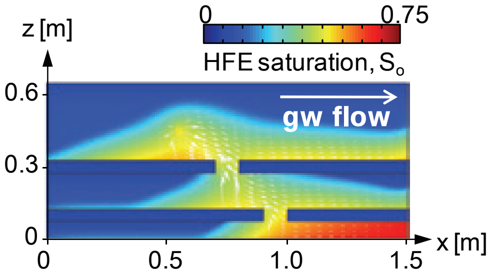
Figure 6 shows the effect of the groundwater flow on the simulated distribution of the HFE. A larger hydraulic gradient affected the HFE migration along the flow direction. More HFE was carried on cracks with a larger hydraulic gradient at a shorter duration. As a result, more HFE infiltrated the cracks in case of a larger hydraulic gradient.



(a) without groundwater flow



(b) hydraulic gradient of 0.033



(c) hydraulic gradient of 0.165

Figure 6. Effect of the groundwater flow on simulated distribution of HFE (using BC model): The flow vectors show the velocity of the DNAPL.

Pump-and-treat remediation is widely used for removal of groundwater contaminated with DNAPLs. The simulation results showed the enhanced groundwater by the pump-and-treat can work not only for removing the groundwater contaminated with the solute of DNAPLs, but also for collecting the source of the DNAPLs around a pumping well. But, it will become difficult to collect the DNAPLs in case that there are some cracks in unconfined aquifers around the pumping well. COMSOL Multiphysics is useful to understand the characteristics and limitations of complex simulation models.

4. Conclusion

This study focused on the displacement pressure effects on DNAPLs migration, and showed the difference between an experimental result and simulation results with two *S-p* models of van Genuchten model, and Brooks and Corey model. The following findings are obtained.

(1) The difference of each model appeared in whether the DNAPLs can permeate unconfined aquifer with relatively low permeability and high displacement pressure or not. The simulation results showed that DNAPLs infiltrated an unconfined aquifer in van Genuchten model, but did not infiltrate it at all in Brooks and Corey model. The simulation result with Brooks and Corey model was almost the same as previous experimental result.

(2) The groundwater flow affected DNAPLs migration. In heterogeneous aquifer including some cracks, a faster groundwater flow enhanced more the velocity of the DNAPLs in the flow direction, but it did not affect the velocity of the DNAPLs in the gravity direction so much.

(3) Although pump-and-treat for removing contaminated groundwater is also effective for collecting DNAPLs around a pumping well, it becomes difficult to collect the DNAPLs in case that there are some cracks in unconfined aquifers.

COMSOL Multiphysics can easily customize the model. It will be expected that simulation models with a high accuracy are increasingly developed through the comparison between the experiments and the simulations.

5. References

1. Kamon, M. Endo, K. Kawabata, J. Inui, T. and Katsumi, K., Two-dimensional DNAPL migration affected by groundwater flow in undconfined aquifer, *Journal of Hazardous Materials*, **110**, 1-12, (2004)
2. van Genuchten, M.T., A closed-form equation for predicting the hydraulic conductivity of unsaturated soils, *Soil Science Society of America Journal*, **44**, 892-898, (1980)
3. Brooks, R.H. and Corey, A.T., Hydraulic properties of porous media, *Hydrology Paper no.3*, Colorado State University, Fort Collins, 1-27 (1964)