CHAPTER 2
LITERATURE REVIEW AND THEORETICAL BACKGROUND

2.1 NAPL model development

Multiphase contaminant transport has been investigated by many researchers. At first, finite difference methods were applied to simulate NAPL transport problems. For example, Kueper and Frind (1991) developed a two-phase flow (oil and water) finite difference model. DNAPL transport patterns in saturated porous media were studied in this work. The finite difference equations were solved fully implicitly using Newton-Raphson iteration and formulated in terms of wetting phase pressure and wetting phase saturation. The results were validated by comparing with one-dimensional analytical solutions and a parallel-plate laboratory experiments.

A three-dimensional, three-phase flow (water-oil-air) finite difference method was developed by Sleep and Sykes (1993). Sleep and Sykes (1993) developed the model by including several numerical options, ranging from fully implicit with first-order upstream weighting to implicit in pressure, explicit in saturation and concentration with third order upstream weighting. This work also compared accuracy and time efficiencies between different numerical approaches.

STOMP (Subsurface Transport Over Multiphase Phases,) is another three dimensional, three-phase, finite difference model. White et al. (1995) developed STOMP to simulate multiphase flow problems using a constitutive equation that included hysteretic imbibition and drainage saturation paths, fluid entrapment, and dissolution of entrapped NAPL.

Finite element method is one of the most efficient methods to solve non-linear problems. Because finite difference methods have low accuracy and are difficult to apply for complicated geometries, many researchers tend to develop NAPL transport models by using the finite element methods.

For example, Kuppusamy et al. (1987) developed finite element model for multiphase flow through soil involving three immiscible fluids: air, water and NAPL. By assuming constant air phase pressure, a variational method is employed for finite element formulation corresponding to the coupled differential equations governing the flow. The model was validated successfully in 2-D using a field study of a leaking underground storage tank. Kaluarachchi and Parker (1989) further developed Kuppusamy et al. (1987) model. One- and two-dimensional simulations were performed to evaluate the accuracy and efficiency of the various schemes (Picard and Newton-Raphson nonlinear iteration schemes) with respect to nonlinear soil properties, time marching, mass balance errors, and soil heterogeneity.

While NAPL transport models are widely studied, the problem with soil deformation, which is an important engineering problem, received very little attention. Shrefler et al. (1994) was the first to combine multiphase transport phenomena and deformation of the porous medium. This is very important when studying the effects of removing fluids from the subsurface. Rahman et al. (1999) further developed Shrefler et al. (1994) model to describe the flow of multiphase immiscible fluids in deforming porous medium. The governing equations describing the displacement of soil and multiphase fluid pressures are coupled together and the resulting nonlinear partial differential equations were solved by the finite element method. Nonlinear saturation and relative permeability functions were incorporated into a Galerkin finite element model to simulate multiphase systems and subsurface migration of NAPL contaminant in porous media. Both the above described works were developed based on Li and Zienkiewicz (1992) theory, which was derived based on the assumption that the gas phase pressure remained constant and equal to ambient air pressure.

Muraleetharan and Wei (1999a) developed a finite element computer code, called U_DYSAC2, for static and dynamic analysis of saturated and unsaturated porous media problems. Three-phase analysis (soil, water, and air) is used to explain the behavior of unsaturated porous media through the Theory of Mixtures with Interfaces (TMI; Muraleetharan and Wei 1999b; Wei 2001; Wei and Muraleetharan 2002a, 2002b). Many researchers have shown that the interfaces between various pore fluids play a major role in the behavior of multiphase porous media. In contrast to conventional theory of mixtures, in TMI the interfaces existing between various bulk phases are explicitly considered. In TMI air pressure changes are also calculated rigorously. U_DYSAC2 is one of the newly developed computer codes to solve for many kinds of problems, for example: wave propagation, two- and three-phase flow and consolidation (deformation of the soil skeleton), and earthquake loading of unsaturated soil embankments.

This project is aimed at extending the capabilities of U_DYSAC2 by modifying U_DYSAC2 to support subsurface contaminant transport problems.

2.2 Experimental investigation of DNAPL transport through subsurface

Many researchers have conducted experimental studies to investigate the NAPL transport through subsurface. Keuper and Frind (1991) conducted 1-g experimental observation of multiphase (DNAPL and water) flow through heterogeneous saturated porous media. Using a sand pack as the porous medium, Keuper and Frind (1991) studied transport patterns of TCE on a 2-D complex soil structure.

Centrifuge studies have been developed to reduce the experimental cost and time. By increasing the gravitational force, NAPL and water phase can be made to move faster and thereby reducing the time required for experiments. One-g-tests and centrifuge tests have been compared by Comoulos et al. (2000) on DNAPL transport in saturated porous media with inclined layers. The result showed the efficiency of centrifuge modeling to describe NAPL transport.

Two-dimensional centrifuge tests at various gravitational accelerations for perchloroethylene (PCE) were conducted by Spiessl and Tayler (2000). The tests showed the instability of the plume's migration, which came from many effects, for example PCE's physical properties (high density, low viscosity), entrapped air and variations in soil properties. DNAPL transport patterns for several gravitational acceleration values were, however, successfully obtained in this study.

Pantazidou et al. (2000) and Abu-Hassanein & Pantazidou (1998) studied the infiltration of high viscosity low density DNAPL in a saturated soil. This study showed that distribution of high viscosity and low density DNAPL was stable and resulted in smaller contamination areas of high DNAPL saturation when compared with a low viscosity and high density DNAPL.

2.3 Constitutive relationships for NAPL-water systems

2.3.1 Capillary pressure and degree of saturation

When two fluids are in contact, a curved surface will tend to develop at the interface and the difference between pressures of two phases is called the capillary pressure.

$$P_C = P_N - P_W \tag{1}$$

Where, P_N = pressure in the non-wetting fluid, and P_W = pressure in the wetting fluid.

In NAPL-water system, capillary pressure is the difference between the NAPL phase pressure and the water phase pressure

$$P_C = P_{NAPL} - P_W \tag{2}$$

For a given porous medium, there are many experiments that try to explain the relationship between capillary pressure and degree of saturation. A typical such relationship is shown Figure 3.1.

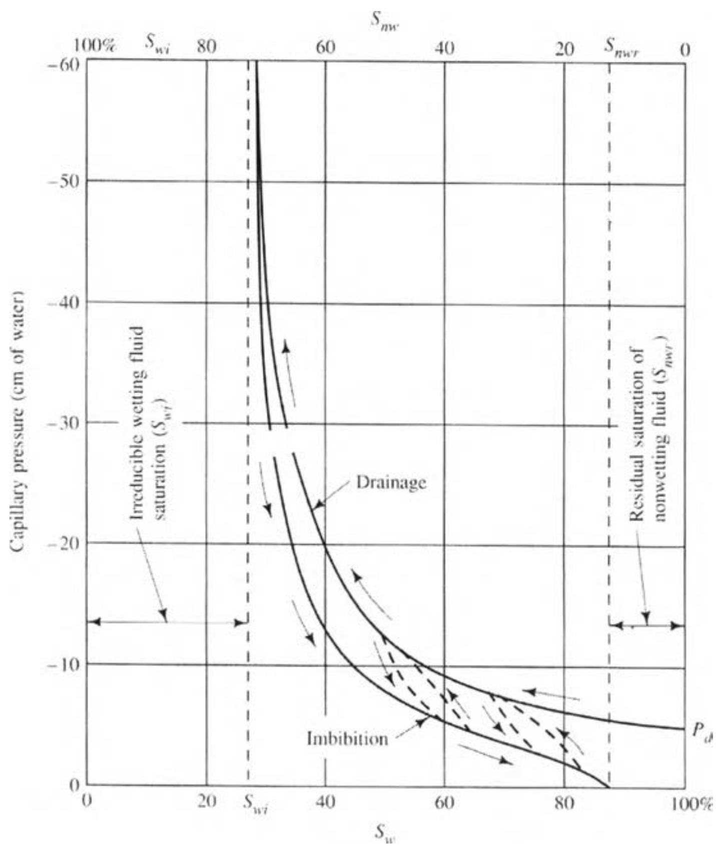

Figure 2.1 : Capillary pressure-wetting fluid saturation curve for a two fluid system, Fetter, C.W. (1992). Contaminant Hydrology

If medium starts off saturated with wetting fluid and the non-wetting fluid displaces wetting fluid, the curve is called the "drainage or drying curve". If the wetting fluid displaces the non-wetting fluid in a medium that is saturated with nonwetting fluid, the curve is called the "imbibition or wetting curve". The imbibition curve does not follow the same path of the drainage curve , this phenomena is called hysteresis.

There are many works that try to explain the hysteresis behavior. The complexity of the hysteresis makes it difficult to simulate the behavior of two phases. To avoid this problem, typically the assumption is made that NAPL contamination problem is on the drainage curve. This is a reasonable assumption for a general environmental problem such as the leaking underground storage tank that contaminates initially water saturated subsurface.

In U_DYSAC2, Brook and Corey (1964) equation is used to explain relationship between effective water saturation (S*) and capillary pressure (Pc). Brooks and Corey (1964) conducted fairly accurate predictions with their equations (equation 3). But this equation still has a discontinuity in the slope at some suction pressure (this point is often referred as bubbling pressure, P_b). This equation creates numerical difficulties when solving purely flow problems due to the use of bubbling pressure in the equation.

$$S^* = \left(\frac{P_c}{P_b}\right)^{-\lambda} \tag{3}$$

where $S^* = \frac{S - S_{irr}}{1 - S_{irr}}$

S^*	= Effective water saturation
S	= Degree of saturation
S_{irr}	= Irreversible degree of saturation
Pc	= Suction pressure or capillary pressure
Pb	= Bubbling pressure
λ	= Pore size distribution index

In modified U_DYSAC2 for NAPL flow problems, the van Ganuchten (1980) equation is used instead of Brook and Corey (1964) equation. Parker et al. (1987) developed saturation–capillary functions for NAPL based on van Genuchten's equation (1980) as,

$$S^{*} = \left[1 + (\alpha P_C)^n\right]^{-m} \quad \text{when} \quad P_C > 0 \qquad (4)$$

where $S^{*} = \frac{S - S_{irr}}{1 - S_{irr}}$ and $m = 1 - \frac{1}{n}$

S^{*} = Effective saturation

S = Degree of saturation

S_{irr} = Irreversible degree of saturation

So,

$$S = S_{irr} + (1 - S_{irr})\left[1 + (\alpha P_C)^n\right]^{-m} \quad \text{when} \quad P_C > 0 \qquad (5)$$

and $$S = 0 \quad \text{when} \quad P_C \le 0$$

Parker et al. (1987) obtained parameters for the main drainage curve for sandy and clayey porous media through nonlinear least squares fitting of experiment data. These values are shown in Table 2.1.

Table 2.1 : Parameters for multiphase saturation pressure function for sandy and clayey porous media (after Parker et al. 1987)

Parameters	Sand	Clay
α_{aw}	0.052	0.032
α_{ao}	$0.099^{(1)}$	$0.108^{(2)}$
α_{ow}	$0.110^{(1)}$	$0.060^{(2)}$
n	1.84	1.86

*α_{aw} = *air-water system,* α_{ao} =*air-oil system,* α_{ow} =*oil-water system*

(1) p-cymene is used for oil phase/

(2) o-xylene is used for oil phase.

Figure 2.2 shows relationships between capillary pressure and degree of saturation using the parameters obtained by Parker et al. (1987)

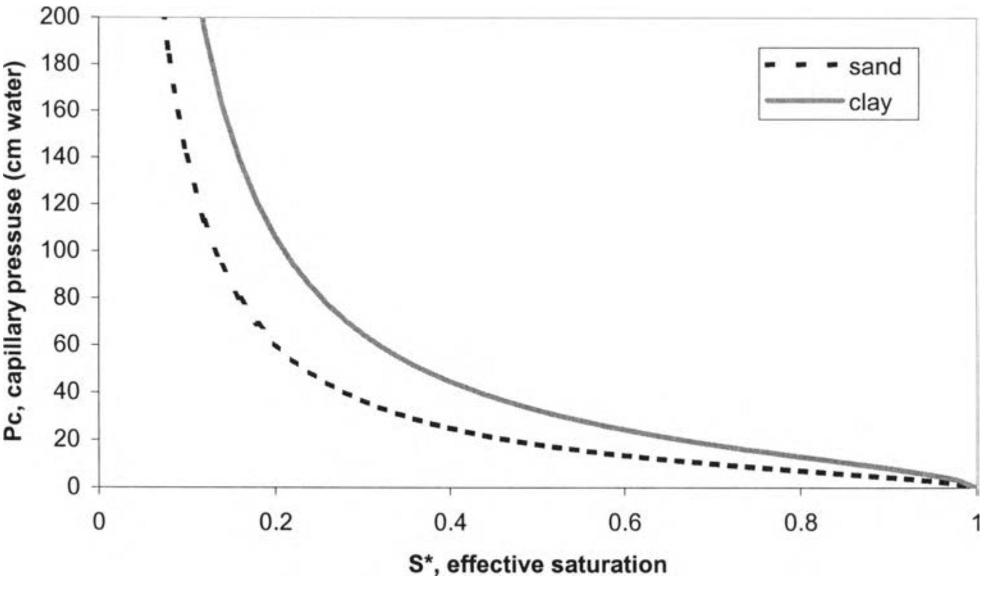

Figure 2.2 : Capillary pressure versus effective water saturation curve

2.3.2 Relative permeability and degree of saturation

The permeability of a fluid varies with the volumetric content of that fluid. In a NAPL-water system, the permeability of the medium with respect to any individual fluid is lesser than when the pore space is fully saturated by one fluid. The reduction in permeability depends on the effective saturation of that fluid, and is described in term of relative permeability, $k_{r\alpha}$ which can be defined as,

$$k_{\alpha}(S^{*}_{\alpha}) = k_{r\alpha} * k_{s\alpha} \tag{6}$$

where $k_{\alpha}(S^{*}_{\alpha})$ = permeability of soil with respect to phase α at effective saturation (S^{*}_{α})

$k_{s\alpha}$ = permeability of soil at complete saturation with phase α

$k_{r\alpha}$ = relative permeability at effective saturation S^{*}_{α}

Parker et al. (1987) developed functions to describe the relationship between effective saturation and relative permeability as shown,

$$k_{rw} = S^{*1/2}\left(1 - \left[1 - S^{*1/m}\right]^{m}\right)^{2} \tag{7}$$

$$k_{rN} = (1 - S^{*})^{1/2}\left(1 - S^{*1/m}\right)^{2m} \tag{8}$$

From equations (7) and (8), it can be seen that at fully water saturated system $S^{*}_{w}=1$, $k_{rw}=1$ and $k_{rN}=0$. When the NAPL starts displacing water k_{rN} will increase and k_{rw} will decrease. When the medium is filled completely with the NAPL, k_{rw} will be equal to 0 and k_{rN} will equal to 1. Figure 2.3 shows the relationships between relative permeability and effective saturation based on Equations (7) and (8)

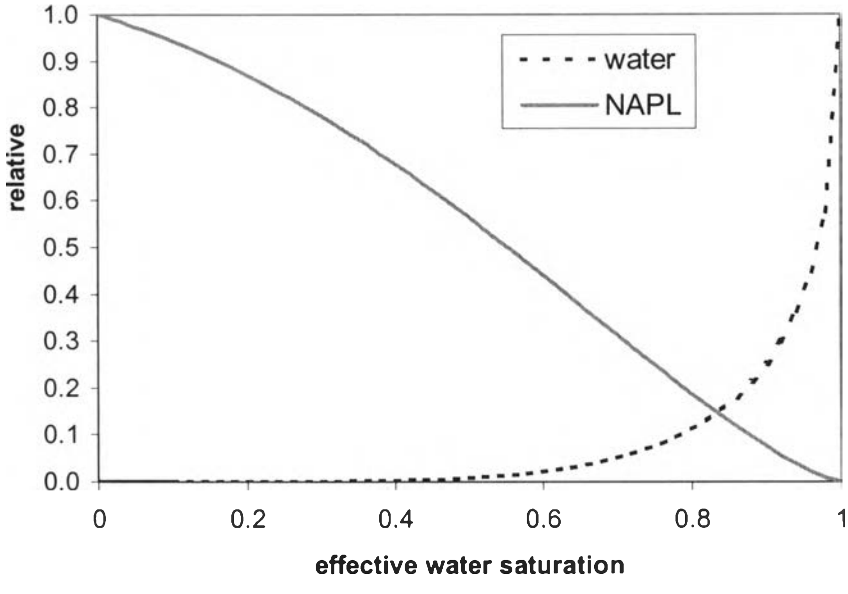

Figure 2.3: Relative permeability versus effective water saturation curves