Migration of Benzene as Light non-aqueousPhase in the Stratified Soil under Unsaturated Condition

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Abstract

A COMSOL Multiphysics software depended on finite element numerical solution is used for prediction the spatially and temporally propagation of the benzene front as non-aqueous phase liquid through the stratified soil consisted of clay and Kerbala's sand as domain (1.3*2) m. The variation of many parameters such as capillary pressure, benzenedistribution, trans missive properties of the soil.Thedegree of saturation as a function of capillary pressure and relative permeability during unsaturated conditions were determined.

The results showed that the benzene front in the Kerbala's sand with presence of clay retained a regular, circular shape at the initial stage of the spill, shortly after that it become ellipsoid as it was advancing. The maximum saturation occurred below the source of the contaminant during infiltration stage. All analysis showed that the presence of clay layer controls the vertical movement of $NAPL_s$ in heterogeneous porous medium.

Keywords:*unsaturated zone, capillary pressure, non-aquos phase liquid, Saturation.*

الخلاصة

تم استخدام برنامج كمسول الذي يعتمد على العناصر المحدده كحل عددي لمعادله انتشار ملوث البنزين في تربه متعدده الطبقات في المنطقه الغير المشبعه.تم اعداد نموذج رياضي يصف حركه الملوثات السائله الأخف من الماء المتسربه من خزانات الوقود الى الطبقه السطحيه من التربه ورصد شكل جبهة تقدم الملوث . تم حساب المعاملات رطوبه التربه ومعامل النفاذيه ومعامل التشبع الفعال وكيفيه تغايرها مع عمق التربه والضغط الشعري . المواد المستخدمة تربه طبيعيه غير متجانسه الرمل والطين كوسط مسامي، والبنزين كملوث. أظهرت نتائج التحليل العددي للعمود التربه (2*1.3) م أن الجبهة الملوث احتفظت بشكل دائري منتظم في المرحلة الأولى من التسرب.تم ايجاد التغاير بين درجه التشبع مع الضغط الشعري و النفاذيه لتربه سطحيه غير مشبعه.

الكلمات الداله: المنطقه الغير المشبعه ،الظغط الشعري،السوائل الأخف من الماء،التشبع.

1.Introduction

Groundwater is one of the most widespread sources of water, and because of its extensive use, groundwater contamination has become a major environmental concern. The great majority of groundwater contaminants are released either from leaking hazardous waste landfills and hazardous waste ponds, or from spills and leaks during the storage and transportation of Non-Aqueous Phase Liquids ($NAPL_S$) on the surface soil as shown in fig.(1). $NAPL_s$ are long-term sources for continued groundwater contamination at many sites (Osborne, 1986) and common pollutants for groundwater supplies because of their widespread production utilization and disposal. $NAPL_s$ have been divided into two general categories; dense and light. Dense Non-Aqueous Phase Liquid (DNAPL) has a specific gravity greater than that of water, whereas the specific gravity of Light Non-Aqueous Liquid (LNAPL) is less than that of water. LNAPL such as Gasoline, Benzene penetrates the soil, it eventually may reach the groundwater table and begin floating on the water surface. it has the potential of migrating with the ambient groundwater velocity, there by contaminating the soil and drinking supply.

The unsaturated zone is a three fluid phase system (water, $NAPL_S$. and air) but in the derivation of the model, air was treated as an immobile phase at constant atmospheric pressure. Oil properties such as density, viscosity, interfacial tension, solubility and vapor pressure are important in understanding oil transport and in predicating subsurface contamination.

Generally, the study of water flow in unsaturated zone comes implicitly with thedevelopment of the flow equation that describes the water movement in a saturated zone. In 1856, Henry Darcy as cited by (Bear, 1972) investigated the flow of water in vertical homogeneous sand. Richard's equation is the commonly accepted for detailed studies of soil water movement, but the computational penalties and apparent difficulties of obtaining the required soil hydraulic properties.Moridis,et al., (

1993)produced analytically based solutions of Richard's equation for restricted conditions. The numerical solution of Richard's equation is becoming more attractive. Ross, (1990) have presented efficient finite-difference solutions of Richard's equation for homogenous soils below the air entry potential.Wipfler, (2003) developed a simple collection of integrated models called the "Hydrocarbon Spill Screening Model" to help in predicted the environmental impact on groundwater from LNAPL spills. Al-Dulaimi (2006) studied the infiltration and redistribution of Light Non-Aqueous Phase Liquid for the state of three fluid phases (water, oil and air) in the unsaturated-saturated zone of the soil. The results showed that the oil plume front, it becomes irregular, characterized by fingering and more extensive lateral than vertical movement. The movement of ($NAPL_S$) in unsaturated-saturated zone of heterogeneous soil and the modeling of variably saturated flow are notoriously difficult to parameterize because several material and hydraulic properties change values as the pressure and saturation levels fluctuate transportation.Theobjectives of this research are to investigate the migration and distribution of LNAPL released in the unsaturated zone above un confined aquifer , and to estimate the volume and distribution of LNAPL plume in the subsurface, resulting from oil spillage in the heterogeneous soil at specified times with different types of boundary conditions.Comsolmultiphysics used to solve the two-dimensional, three-phase flow problemsin the heterogeneous soil.

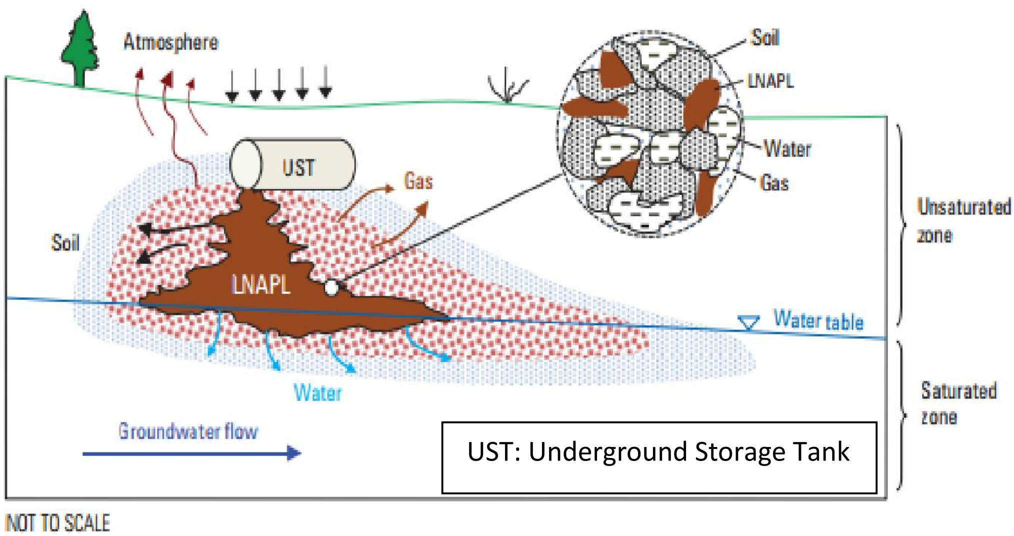


Fig.(1): Containment of $NAPL_s$ migration by horizontal subsurface barrier (Wonyong Jang,2013)

2. Materials

A- Contaminant Liquid

Benzeneis an important hydrocarbon compounds. it was chosen as the $NAPL_S$contaminant because of its high health hazard. Also it has some desirable properties such as specific gravity lower than 1.0, very low solubility in water. Table (1) summaries the most important properties used in the present research.

Table (1): Benzene properties used in the present research.

Parameter	Value
Specific gravity	0.8765 at (25 °C)
Viscosity	0.6076 cP (25 °C)
Chemical formula	C6H6
Solubility in water at (25 °C)	1.8 g/L
Appearance	Color Less
Main hazard	carcinogen, flammable

B- Soil

Natural Iraqi soil used as porous medium. The porous mediums used were sand as lower layer and clay as top layer,sand is known as "Kerbala's sand". The sand, clay was clean and tested in soil lab to obtain the characteristics of the soil. Table (2) summaries the properties of this soil.

Table (2): The characteristics of the soil

Property	Sand	clay
Particle size distribution (ASTM D422) Effective size D_{10} (mm) Mean grain size D_{50} (mm) Uniformity coefficient Cu Gradation coefficient Cc	 0.18 0.36 2.34 0.826	 P.I=56
Coefficient of permeability (m/day) @ 20°C (ASTM D2434-68).	0.4501	.0212
Organic content (%) (ASTM D 2974)	1.1	2.6
Porosity (*n*)	0.386	.66
Soil classification (ASTM D422)	Uniform sandy soil	Clay Soil with High Plasticity

3. Governing Equations

The Richards' equationis governs fluid flow in variably saturated porous media.The equation is

$$[C+S_e\, S]*\frac{\delta Hp}{\delta t} + \Delta\ [-k_s k_r \Delta(Hp+D) = Q_s \ldots\ldots\ldots\ldots\ldots(1)$$

where **C**is specific moisture capacity (m^{-1}); **Se** is the effective saturation; **S**denotes the storage coefficient (m^{-1}); **H*p***equals the pressure head (m); *t* is the time (day); **K*s***represents the hydraulic conductivity ($m^{-1}\cdot day^{-1}$); ***kr***gives the relative permeability; **D** is the coordinate (for example *x*, *y*, or *z*) for the vertical elevation; and ***Qs*** represents the fluid source defined by volumetric flow rate per unit volume of soil (day^{-1}). Here **S**equals the difference between the liquid volume fraction at saturation, **θ*s***, and the residual liquid volume fraction, **θ*r***, or specific yield per unit length.

Changes in pressure head and elevation head drive fluid through the soil**. *K*, θ, *C***, and **Se** vary under unsaturated conditions (***Hp*< 0**), and they reach a constant value when the system saturates (***Hp*≥ 0**).Where pressure head at saturation is atmospheric (*Hp*= 0).The parameterization is as follows:

$$\Theta = \begin{cases} f(Hp) & Hp < 0 \end{cases} \quad \text{------------------(2)}$$

$$Se = \begin{cases} \theta_s & Hp \geq 0 \\ (\theta - \theta_r) / (\theta_s - \theta_r) & Hp < 0 \\ 1 & \end{cases} \quad \text{-------------------(3)}$$

$$C = \begin{cases} \partial\theta/\partial Hp & Hp < 0 \\ 0 & Hp \geq 0 \end{cases} \quad \text{--------------------(4)}$$

$$Kr = \begin{cases} \frac{K}{Ks} f(Hp) & Hp < 0 \\ 1 & Hp \geq 0 \end{cases} \quad \text{-------------------(5)}$$

4. BoundaryConditions

The boundary conditions are:

$$\begin{array}{ll} H_p = H_{p0} & \partial\Omega \\ \mathbf{n} \cdot [-K_s k_r \nabla(H_p + D)] = 0 & \partial\Omega \ \text{Surface} \\ \mathbf{n} \cdot [-K_s k_r \nabla(H_p + D)] = 0 & \partial\Omega \ \text{Sides} \\ \mathbf{n} \cdot [-K_s k_r \nabla(H_p + D)] = 0 & \partial\Omega \ \text{Symmetry} \\ \mathbf{n} \cdot [-K_s k_r \nabla(H_p + D)] = N_0 & \partial\Omega \ \text{Base} \end{array}$$

The pressure head in the source is constant, and no flow exits in the surface outside the pressure source. The sides are impermeable. The vertical boundary on the inside of the source is a line of symmetry, and the model approximates the small amount of leakage from the base, as being 0.03*Ks*. where**n** is the outward unit normal to the boundary.

Initially, the shallow soil surface has a specified distribution of pressure head within the flow domain, ̓Ω:

$Hp\ (z, r) = Hp_o(z, r)$ at $t=0$

5. Unsaturated Soil Hydraulic Properties

For two-phase system, the unsaturated soil hydraulic properties are determined experimentally and fitted with some empirical mathematical functions of Brooks and Corey as cited by (Kueper and Frind, 1991b). These parameters are the displacement pressure, P_d and the pore size distribution index, λ.

Table(3): Sand, and clay properties used for present research. (Kassim, et al.,2009)

parameters	values
sand displacement pressure. cm (P_{dh})	9.9
clay displacement pressure .cm (P_{dl})	36
sand residual wetting phase %(S_{rh})	9
clay residual wetting phase % (S_{rl})	10.5
sand pore size distribution index (λ_h)	1.343
clay pore size distribution index (λ_l)	1.25

6. Model Definition

The field contaminant transport from a waste disposal site into a unconfined aquifer is consist of towlayers . Top layer is clay have athickness of (0.5) m and the lower layer is sand have a thicknessof (.8) m. The source of contaminant (oil) of (0.3) m radius sits at the ground surfaceand infiltrates from the disposal site into the unsaturated zone under (1)cm head constant conditions**.**The source is bottomless, so oil moves from the source into the soil. Thewaste disposal site itself has lateral dimensions of a(2) m long and soil column of radius (1.3) m. Transient flow problems are solved by time marching until a prescribed time is reached. The finite element mesh is constructed by dividing the flow region into tetrahedral prismatic elements as shown in fig.(2).The variably saturated flow problemis solved using

(ComsolMultiphysicsV 3.5a)assuming an iteration tolerance of 0.001.

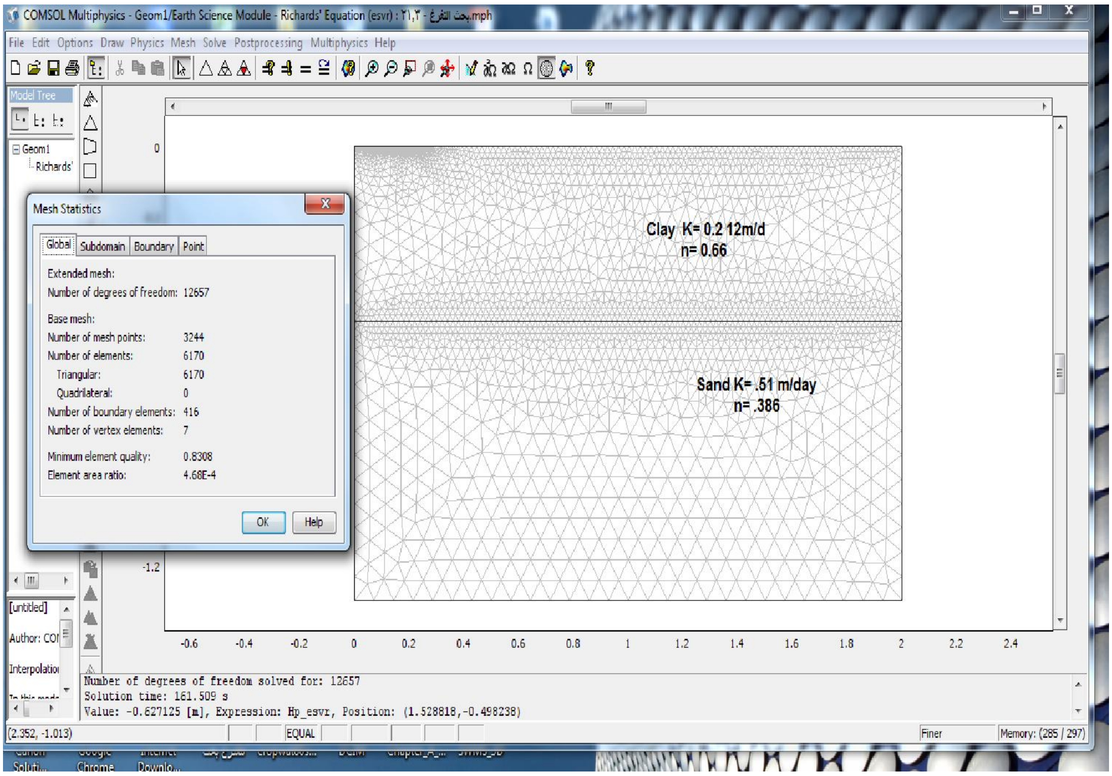


Fig.(2): Domain discrezation of unconfined aquifer.

7. Results and Discussion

7.1Pressure Head Distribution

Fig.(3) shows the distribution of capillary pressure in unsaturated zone for two layers after 10 days from benzene spill.

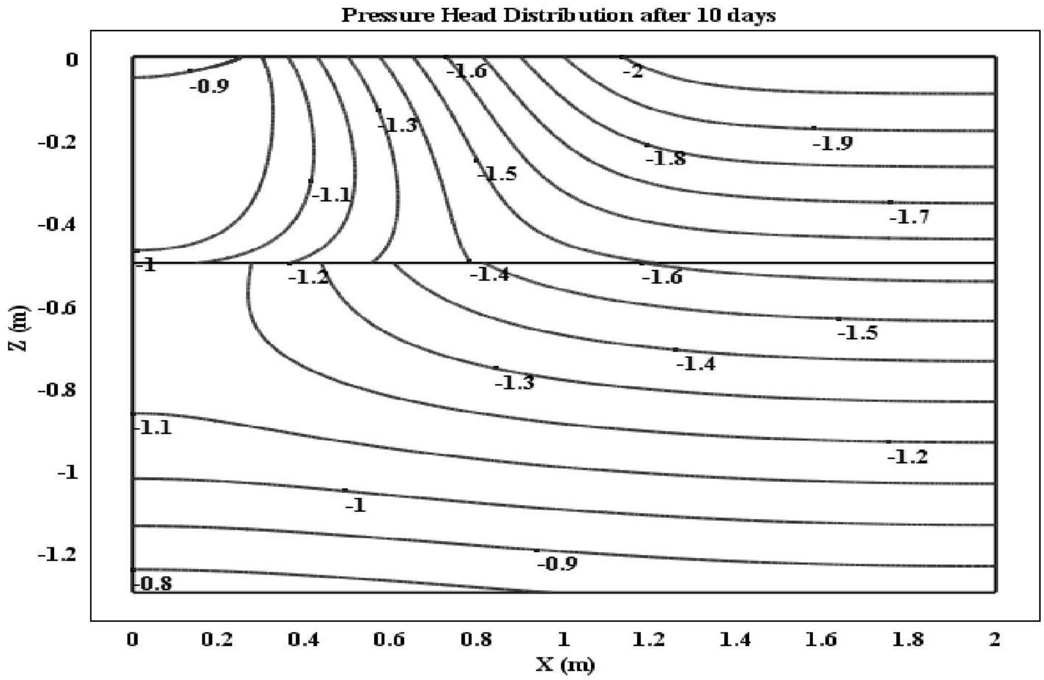
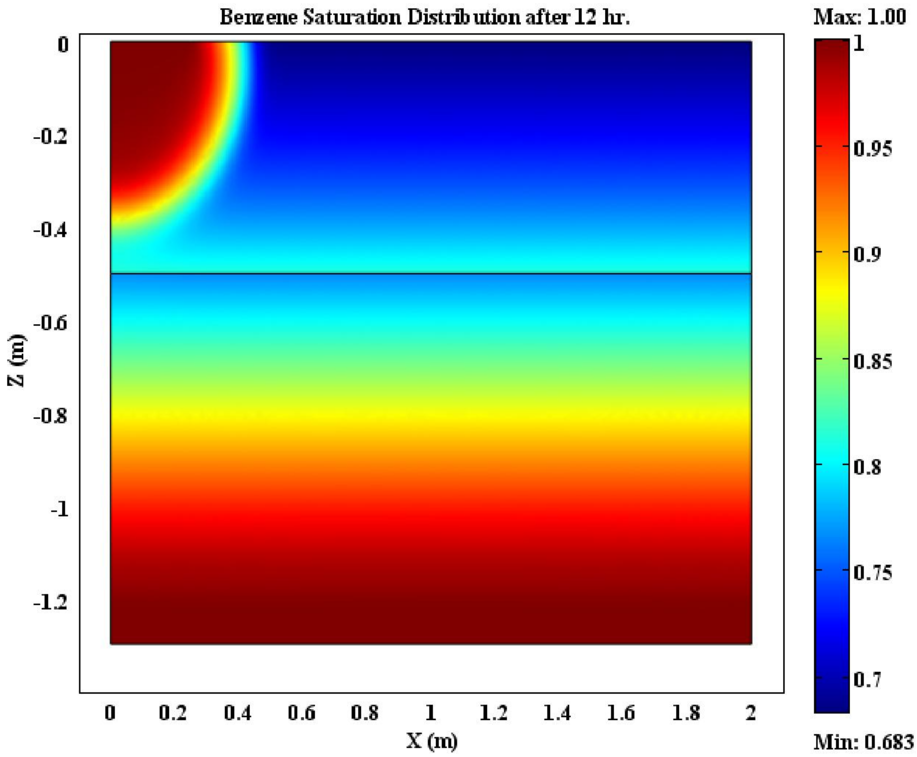


Fig.(3): Capillary pressure after 10 days from spill.

7.2 Propagation of Oil Plume

The propagation of the plume is observed at appropriate intervals during infiltration above the two layers. The visual observation of the spread of benzeneplume after (12 hr.) and (48 hr.) of infiltration is depicted in Fig.(4)



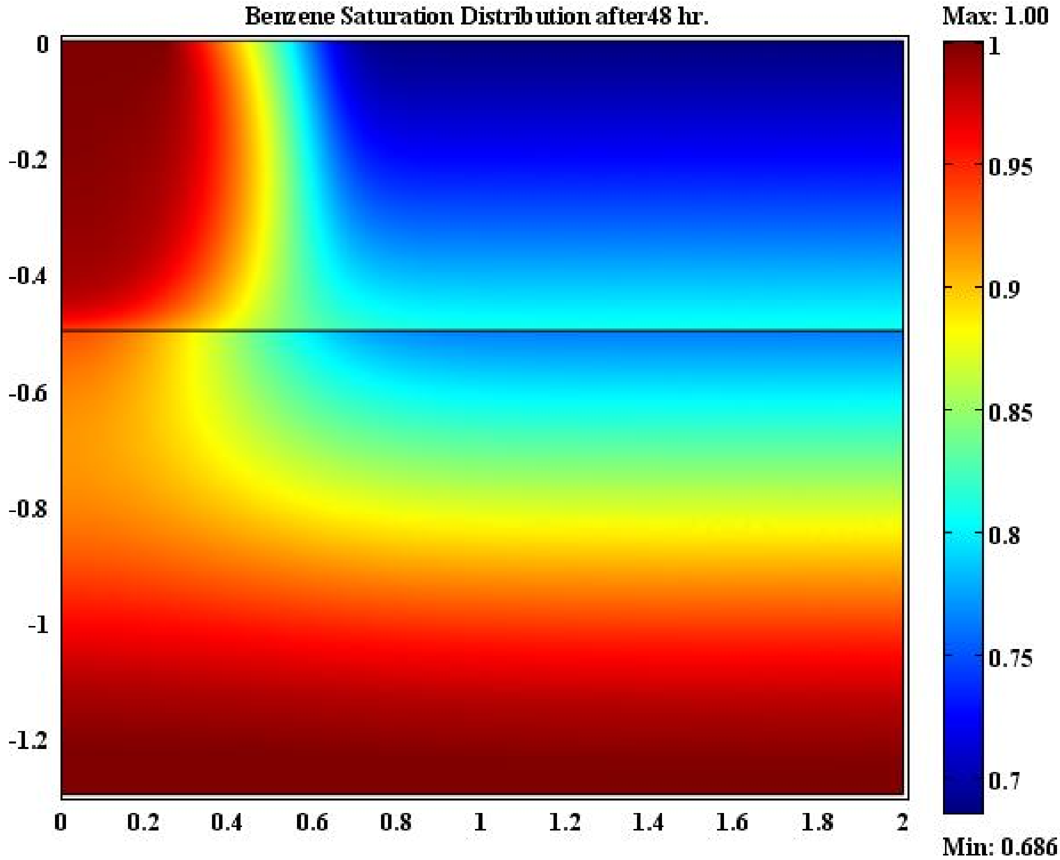


Fig.(4): Benzene distribution a. after 12 hr. b. after 24 hr.

7.3 EffectiveSaturation with Depth

Fig. (5) shows benzene saturation distributions curves at different time. It is obvious that the range of benzene saturation is changed evidently with increasing time. The saturation of clay soil greater than sand soil.

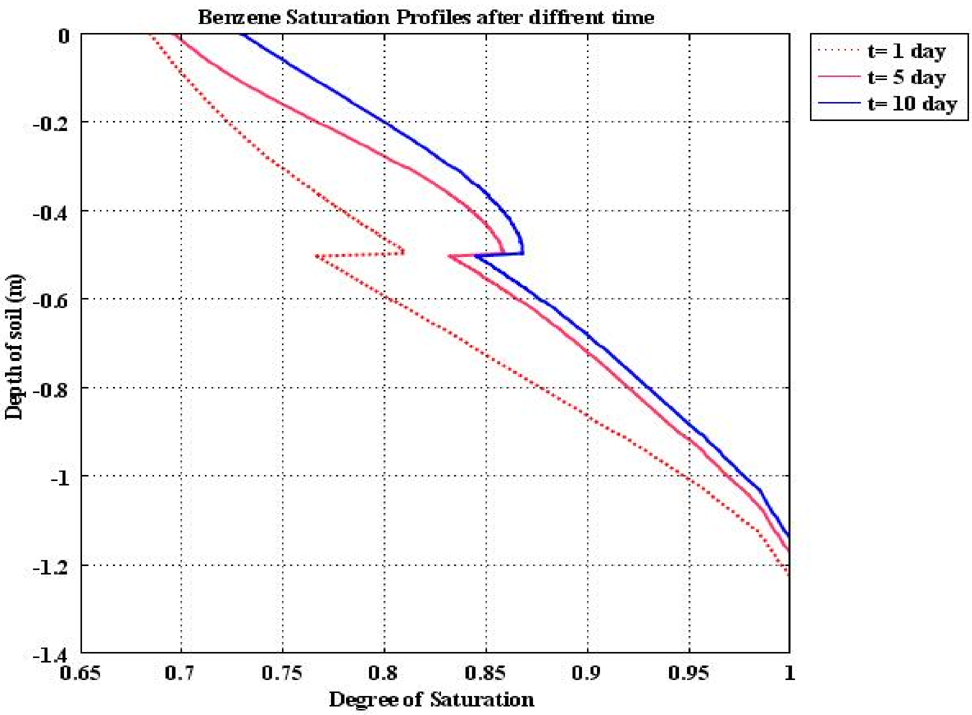


Fig.(5): Benzene Saturation after different time

7.4 Hydraulic Conductivity with Depth

Fig. (6) Show benzene hydraulic conductivitycurves at different time. It is obvious that the range of conductivity was changed slowly with increasing time. The permeability of clay soil less than sand soil.

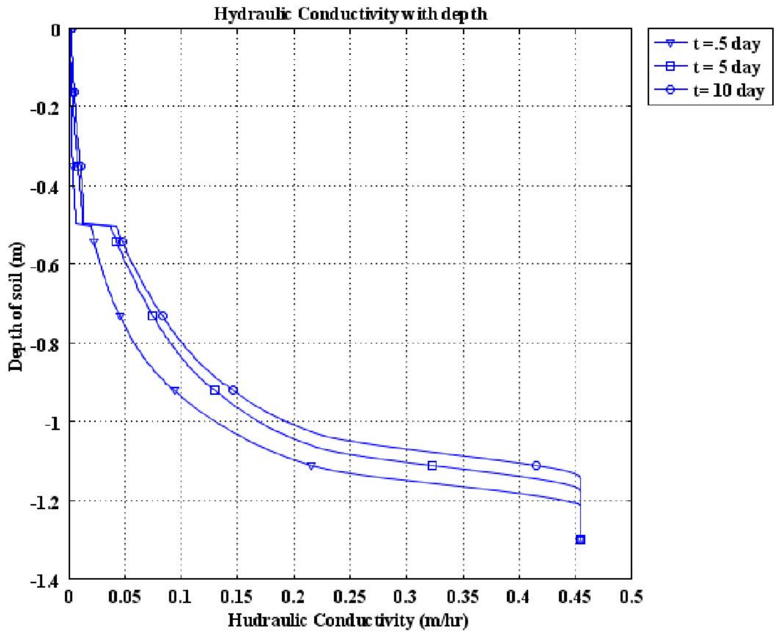


Fig.(6): Hydraulic conductivity after different time.

7.5 Capillary Pressure with Degree of Saturation

Fig.(7) shows the relationship between capillary pressure with benzene saturation.This relation after 10 days from benzene spill.

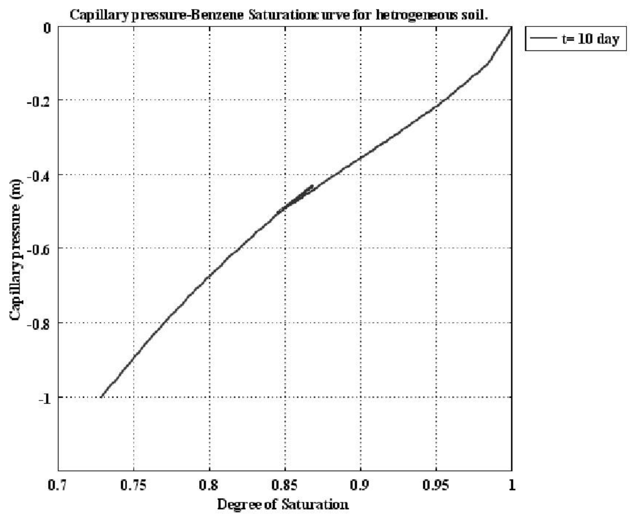


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Fig.(7): Capillary pressure –benzene saturation for heterogeneous soil.

7.6 Relative permeability with Degree of Saturation

Fig. (8) shows the relation between relative permeability with degree of saturation after 10 days from benzene spill.Permeability change rule under the unsaturated flow and sand permeability rabid allyincrease with the increasing of benzene saturation. It is obviously that changes in 0.85 ~ 1 range in saturation.in clay layer the slowly increase of relative permeability with the increase benzene saturation in ranges 0.73 ~ .87.

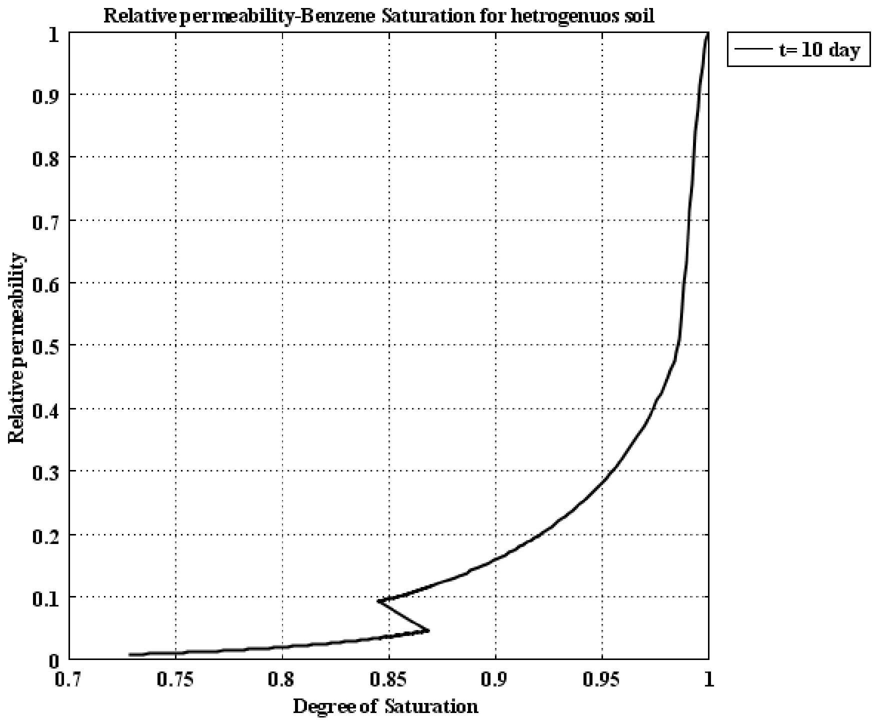

Fig.(8): Relative permeability-Benzene saturation curve of heterogeneous soil.

8. Conclusions

The numerical solution of the governing equations forRichards' equation with Comsol techniques showed to be an efficient procedure in solving two- dimensional Light Non-Aqueous PhaseLiquid(LNAPL) flow through unsaturated zone in three phases system. The plume was traced at appropriate intervals. During the vertical migration of plume at the clay layer of infiltration, the benzene plume front retained a circular shape, after 24 hr. approximately the front shape became ellipsoid as it advanced but later on, as it reached the sand it became irregular with lateral spreading above the sand although some of (LNAPL) penetrated the clay. This spreading continued at faster rate because of higher relative permeability of the sand compared with lower relative permeability of the clay.

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