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Non-aqueous phase liquids distribution in three-fluid phase systems in double-porosity soil media: Experimental investigation using image analysis

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ABSTRACT

Over the last few decades, contamination of groundwater and soil by non-aqueous phase liquids (NAPLs) has become a serious and wide-spread problem for the environment In this research, a light transmission visualization (LTV) method was used to observe the migration of dense non-aqueous phase liquid (DNAPL) and light non-aqueous phase liquid (LNAPL) in double-porosity soil within a three-fluid phase system (air-NAPL-water). The double-porosity characteristics of the soil were created using a composition made up of local sand and sintered kaolin clay spheres arranged in a periodic manner. Toluene was used to simulate LNAPL while tetrachloroethylene (PCE) represented the DNAPL. Both NAPLs were dyed using Oil-Red-O for better visualization. For comparison purposes, the same experiments were carried out using just local silica sand acting as a type of single-porosity soil. A significant difference in the migration of the toluene and PCE was observed as both the NAPL migration rates in the double-porosity medium were much faster compared to the migration rates found in the single-porosity medium. This result is most likely due to the occurrence of inter-aggregate pores in the double-porosity soil that contribute to increasing velocity of fluids migration through porous media. Other factors such as the wettability of fluids and capillary pressure characteristics that exist in the soil pores were found to be influential factors in fluid migration within porous media. In addition, the results show that chemical properties have a significant influence on the NAPL migration in porous media. It was found that the migration velocity of toluene was much faster compared to the migration velocity of the PCE. This observation is most likely caused by the fact that the distribution coefficient of toluene was higher than that of PCE which in turn means that the retardation factor of toluene is lower than that of PCE in the same porous media. This paper proved that the LTV provides a non-intrusive and non-destructive technique for studying multiphase flow in double-porosity soil media where rapid changes in fluid distribution in the entire flow domain is not easy to measure using conventional tools.

1. Introduction

Contamination of groundwater and soil by non-aqueous phase liquids (NAPLs) has become a serious and wide-spread problem for the environment (Luciano et al., 2012; Tick et al., 2015). Spills of petroleum products and leaks from underground storage tanks are examples of NAPL releases in the subsurface system. Generally, NAPLs are divided into two main types based on its density; dense non-aqueous phase liquid (DNAPL) which is denser than water and light non-aqueous phase liquid (LNAPL) which has density less than water. Chlorinated solvents such as trichloroethylene and tetrachloroethylene are the most spread examples of DNAPL, whereas benzene, toluene, ethyl-benzene and xylene (BTEX) are examples of LNAPL (Sweijen et al., 2014). When LNAPL is released into the subsurface, it will migrate downward until it reaches the water table (Mercer and Cohen, 1990). For DNAPL, it is behaviour is similar to LNAPL in the unsaturated zone after being released into the subsurface but while LNAPL tends to pool on groundwater it encountered, DNAPL will continue to

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migrate downward vertically into the saturated zone (Ngien, 2012). It is very important to understand the migration of NAPLs in subsurface systems in order to evaluate the contamination source zones as well as to design relevant remediation schemes (Zheng et al., 2015). Investigation of contaminant hydrological processes often relies on indirect measurements such as non-invasive techniques (Werth et al., 2010). Non-invasive techniques can easily investigate the behaviour of fluids flow in the soil samples without disturbing the samples such as X-ray, gamma ray and photographic methods (Alazaiza et al., 2015). A substantial number of experimental studies have been conducted to investigate the behaviour of immiscible fluids in soils (Alazaiza et al., 2017b; Pan et al., 2016; Zheng et al., 2015; Sa'ari et al., 2015; Ngien et al., 2012; Bob et al., 2008; Darnault et al., 1998; Kechavarzi et al., 2005). All these studies were carried out to investigate the behaviour of NAPL in one or two-dimensional, two-phase (NAPL-water) systems. A few experimental studies on NAPL migration in three fluid-phase systems were found but these were limited to sand being used as the single-porosity media (Darnault et al., 2001; Kechavarzi et al., 2000; Schroth et al., 1998).

In the last few years, several non-destructive and non-intrusive methods used to observe fluid saturation, such as X-ray attenuation (Tidwell and Glass, 1994) and gamma-ray techniques (Høst-Madsen and Jensen, 1992) have been reported. However, these techniques do not allow the observation of dynamic fluid saturation in the whole flow domain at the same time. Other limitations include, long counting times as well as only one point can be acquired at one time (Darnault et al., 1998). Due to the mentioned limitations, light transmission visualization (LTV) technique has been gained more attention by several researchers (Alazaiza et al., 2016; Bob et al., 2008; Niemet and Selker, 2001; Zheng et al., 2015). A review by Alazaiza et al. (2015) presented the usage of three types of photographic methods in NAPL experiments and showed that LTV is a viable method in NAPL saturation measurements. The theory behind the LTV technique is the passing of electromagnetic energy into the test media where the distribution of liquid saturation is measured as variations in the light intensity field. There is a linear relationship between saturation and light intensity due to closer matching between the refraction index of the porous media and water relative to the porous media and air (Glass et al., 1989). In LTV systems, the change in fluid content and fluid saturation can easily be measured instantaneously due to the utilization of cooled charged coupled device (CCD) cameras which provide a very high density array of spatial measurements over a very large dynamic range (Niemet and Selker, 2001).

LTV was first developed by Hoa (1981) to measure water content in a 2-D sand-filled chamber. After that, Tidwell and Glass (1994) used the LTV technique to measure liquid saturation by correlating the number of pores filled with water to the total number of pores across the thickness of the model as well as to water saturation. Thereafter, Darnault et al. (1998) applied the LTV method to investigate the relationship between hue and water content through full field in a two phase system (water – oil) using the hue, saturation and intensity (HSI) format. They found that the major problem in measuring water and NAPL saturation in the system was due to the similar refraction indices of these liquids. Several years later, Darnault et al. (2001) developed a method to measure fluid content in a three phase system (NAPL –water – air). After a few years, a major contribution to the LTV technique was achieved by Bob et al. (2008) who studied the quantification of PCE saturation in a two dimensional, two fluid phase system in single-porosity silica sand.

Several characteristics influence the migration of contaminants in the subsurface system. One of these characteristics is the soil structure. In the natural state, many types of soils have two distinct scales of porosity which lead to the term double-porosity soil structure (Carminati et al., 2008). Double-porosity is a natural phenomenon (El-Zein et al., 2006) found when two separate pore systems occur simultaneously in the structure of the soil. Rock aquifers (Pao and Lewis, 2002), compacted soils (Romero et al., 1999) and agricultural top soil (El-Zein et al., 2006) are examples of geomaterials that potentially harbour double-porosity characteristics.

The aim of this paper is to study the migration of both LNAPL and DNAPL in double-porosity soil in a 2-D flow chamber in three fluid-phase system (air-NAPL-water) using the LTV technique. To the best knowledge of the authors, studying the migration of both LNAPL and DNAPL in double-porosity soil within a three fluid-phase system using LTV technique has not been done before. The LTV technique was used to qualitatively analyze the migration of both LNAPL and DNAPL in double-porosity soil medium which is useful in designing the remediation methods. To achieve the objective of this paper, two 2-D laboratory experiments were conducted in order to observe the migration of LNAPL and DNAPL in double-porosity soil in three fluid-phase system. For comparison purpose, the same experiments were carried out again but using sand as a single-porosity in three-fluid phase system. Section 2 presents the materials and methods that were used. Section 3 shows the results and the captured images for the behaviour of NAPLs in all soil samples, whereas Section 4 presents the conclusion of the study and possible future work.

2. Materials and methods

2.1. Double-porosity creation

The double-porosity medium was created using solidified clay spheres and fine sand. The solidified clay spheres were made from commercially available kaolin S300 (Lotte Chemical Titan, Malaysia) while the sand used was commercial local silica sand (Lotte Chemical Titan, Malaysia). Physical, chemical and mineralogical properties of the materials were characterized as presented in Alazaiza et al. (2017a).

Double-porosity in the soil samples was created following a previous study by Lewandowska et al. (2005). Sintered clay spheres were made by hand where the kaolin powder was mixed with sufficient amount of water to form the spheres before being burned. The clay spheres were then put inside the furnace and the temperature was increased gradually until it reached 1000 °C. This process was to make the kaolin spheres hard as well as to increase their ability to avoid disintegration in case of saturation. The size of the spheres was almost similar by visually test. The diameter of the sphere was 6 mm. Scanning electron microscopy (SEM) was conducted with different magnification power to investigate the micro-structure of kaolin spheres as shown in Fig. 1. The sand was washed with distilled water to remove all the fine residuals that might be present. As an extra precaution, the washing process continued until the absorption of the waste distilled water, as measured by a spectrophotometer, was similar to the clean rinsing distilled water at the same wavelength. Thereafter, the sand was oven dried for 48 h at 45 °C.

The packing procedure was conducted by pouring the clay spheres layer by layer in such a way that they were touching each other. Between each layer, a 1 cm layer of sand was poured carefully which was then compacted by tapping the outer frame of the model using a plastic hammer. The procedure was repeated to obtain the periodic arrangement. The total porosity of the system was 0.41.

2.2. Experimental setup

2-D flow chambers packed with double-porosity media were used in this research to study the migration of NAPLs in double-porosity media in three-fluid phase system using LTV. These flow chambers were constructed using 10 mm acrylic material with internal dimensions of 45 cm height × 30 cm width × 1 cm depth. The flow chamber was kept in fixed position inside a steel frame fixed onto a stainless steel light box that contained a light source. Two ports attached to the bottom of the flow chamber allowed inflow and outflow of fluids. The ports were connected to a water tank by plastic tube where a valve between the