Estimation of water – LNAPL transfer with different granular materials using X-ray CT image analysis

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ABSTRACT

The objective of this study is to improve the understanding the migration mechanisms of Light Non-Aqueous Phase Liquids (LNAPLs) in porous media at the pore network scale. In this paper, an evaluation of trapped LNAPL in a pore structure after injection experiments coupled with micro focused X-ray Computed Tomography (MXCT) is presented. Image analysis of pore occupation by fluids from MXCT data was performed using different granular materials: glass beads and sandy soil. It can be observed that the trapping process is dependent on the pore structure. Specifically, LNAPL tends to be retained in larger pores in certain area where can work capillary pressure.

Keywords: image analysis, multi-phase flow, ground contamination, X-ray CT scanner

1 INTRODUCTION

Migration mechanisms of Light Non-Aqueous Phase Liquids (LNAPLs) in contaminated soil obey the well-known flow processes: gravity-driven flow and percolation in the unsaturated zone, mostly driven by capillary pressure. Multiphase flow which is water and LNAPL flow in a porous media is dependent on the local structure of the porous medium (pore diameter, connectivity, tortuosity, etc.), on the fluids properties (viscosity etc.) and on the fluid/fluid and fluid/solid interactions (interfacial tension, wettability, etc.). LNAPL may be trapped in the saturated zone due to water level rise, for instance owing to rainfall events. Nowadays, Micro focused X-ray Computed Tomography (MXCT) scanners have resolution of micron unit and allow non-destructive evaluation of phases distribution (both fluid and solid) and morphological parameters such as in the study of fluid behavior in sandy soil. (Wildenschild et al., 2002; Altman, 2005; Wildenschild et al., 2005; Al-Kharusi & Blunt, 2007; Mukunoki et al., 2011). The purpose of this study is to investigate the LNAPL trapping mechanism with different granular materials using MXCT image analysis. This paper introduces an image analyzing technique for evaluating phase’s distribution and pore size. Pore structures trapping LNAPLs are then visualized by cluster analysis, allowing static evaluation of structure in pore scale. Specifications of the MXCT used in this study are in Mukunoki et al. (2011)

2 INJECT EXPERIMENT

2.1 Materials

Two granular materials were used in this study: Toyoura sand with a particle density of 2.64 g/cm³ as the reference soil and glass (SiO₂) beads from Potters-Ballotini Co., Ltd. with a particle density of 2.45 g/cm³. The fluids chosen for the experiment are water and LNAPL. Since water and LNAPL have similar density, distinguishing one phase from another is difficult by CT because CT images are composed of CT values, which are related to material density. Therefore, the water solution was modified by adding potassium iodide (KI) to allow better separation of the CT values. Isoparaffin was chosen for the LNAPL phase. Fluid properties are summarized in Table 1.

Table 1. Fluid properties.

| Property                  | LNAPL | KI solution |
|---------------------------|-------|-------------|
| Density \( \rho \) (g/cm³) | 0.7486| 1.24786     |
| Viscosity \( \nu \) (cP)  | 1.29  | 0.9664      |
| Surface tension \( \gamma \) (dyn/cm) | 20.0 | 72.3        |
| Contact angle \( \theta \) (degree) | 6.4  | 62.1        |
| Interface tension \( \gamma_{nw} \) (dyn/cm) | 54.5 |

2.2 Apparatus and Experiment procedure

Mukunoki et al. (2013) developed the MXCT fluid injection test system used this in the study. Table 2 shows 4 test cases and the granular materials properties. The glass powder means the crashed glass beads. The main experimental steps are as follows:

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1) Saturation of the specimen with KI solution;
2) Injection of 5 pore volumes (PV) of LNAPL with 25 ml/h injection rate;
3) Scanning by MXCT;
4) Injection of 5 PV of KI solution with 500 ml/h (Case2) or 25 ml/h (other cases);
5) Scanning by MXCT.

As for the injection rates of KI solution and LNAPL, in this study, as LNAPL flow in contamination process is considered as slow as the elevation flow of groundwater table, both injection rates are set the same value as KI solution injection rate.

### Table 2. Test cases.

|                  | Case1     | Case2     | Case3     | Case4     |
|------------------|-----------|-----------|-----------|-----------|
| Specimen         | Glass beads | Glass beads | Glass powder | Toyoura sand |
| Grain size (µm)  | 300       | 300 +1000 | 350       | 200       |
| Soil particle density (g/cm³) | 2.45 | 2.45 | 2.45 | 2.64 |
| Dry density (g/cm³) | 1.567 | 1.75 | 1.492 | 1.60 |
| Porosity         | 0.36      | 0.29      | 0.39      | 0.36      |
| Pore volume (cm³) | 5.26     | 4.17      | 5.87      | 5.76      |

### 3 IMAGE PROCESSING ANALYSIS

#### 3.1 Marker-controlled watershed processing

X-ray CT image are composed of voxels, associated with CT values, which are proportional to the material density. The elements observable in the MXCT images in this study are glass beads, Toyoura sand, small amount of magnetic sand, LNAPL and KI. An image treatment is required to separate each component from one another. Figure 1(a) shows an example of CT image with the 4 groups: soil, KI solution, LNAPL and magnetic sand. Knowing the number of materials in the CT images allows fixing areas of magnetic sand particles, soil particles, LNAPL and KI, as well as identifying mixels where the CT value could be attributed to two or more materials. Watershed processing, which is based on CT value gradient changes between materials, was applied only to mixel areas in what is called the marker-controlled watershed (MCW) method. Figure 1(b) shows a post-processed CT image after marker-controlled watershed was performed. The boundary of each material in the CT image with maximum gradient was defined as boundary between different materials, and watershed processing was applied to the CT image. This CT value thresholding technique does not lose spatial geometry information, and is an objective approach for distinguishing the CT threshold values.

### 3.2 Image analysis based on mathematical morphology

Mechanisms of trapping LNAPL are complex because it is dependent on local structure such as pore volume and hysteresis of water retention. Mukunoki et al. (2011) proposed an image processing technique that uses mathematical morphology (Soille, 2002) to evaluate the 3-D distribution of pore size in dry, sandy soil (a two-phase soil particle and air mixture). We used this method to evaluate the volume and diameter of pore filled with LNAPL. Moreover, connectivity of the pore structure affects flow behavior, and so it should be considered. Mukunoki et al. (2013) developed the cluster labeling method. We can apply this method to separate pore blobs, making it possible to evaluate spatial distribution of the LNAPL volume.

### 4 RESULTS AND DISCUSSION

#### 4.1 LNAPL injection test

Figure 2 show results of Case1-Case4 for LNAPL injection test. The results show the evolution over time of cumulative collection of liquids represented by the volume of injected fluid per pore volume at the outlet. The break-through was observed at 0.71PV for Case1, 0.67PV for Case2, 0.60PV for Case3 and 0.65PV for Case4. Final LNAPL saturation is of 71% for Case1, Fig. 2. Result of LNAPL injection.
67% for Case2, 88% for Case3 and 80% for Case4. It can be observed that the final saturation between Case3 - Case4 and Case1 is likely due to grain and therefore pore shape as described in section 4.3. The discrepancy between Case2 and the other cases may be due to both the bimodal grain size distribution in Case2 and the different injection protocol. It should also be noted that the porosity of the materials affect the final saturation in Table 2.

4.2 KI solution injection test

Figure 3 shows the change over time of collection of fluid phase at the outlet. Break-through is reached faster for KI injection than for LNAPL first injection (0.56PV for Case1, 0.52PV for Case2, 0.48PV for Case3 and 0.48PV for Case4). The LNAPL saturation at the end of test is 14.88% for Case1, 17.09% for Case2, 25.61% for Case3 and 21.48% for Case4. The saturated LNAPL is 20.97% for Case1, 25.36% for Case2, 29.05% for Case3 and 26.82% for Case4. From this result, Case3 is the case that allows the most trapped LNAPL.

4.3 Pore size histogram analysis using CT image

Figure 4 (a) shows the MXCT images which are taken after LNAPL injection and KI solution injection for all cases (step 5). MXCT images are generally 256-level grayscale, with black areas having the least density and white areas the greatest. CT image size for image analysis were set at 500×500×500 voxels from center with the same as the total CT image. Figure 4(b) show the 4-color MXCT image created from Figure 4(a) using MCW. Figure 5 show the result of mathematical morphology image processing. The pore shape for Case 3 (Glass beads powder) and Case 4 (Toyoura sand) is constituted by acute shape and Case 1 (Glass beads) is constituted by spherical shape. The difference in shape leads to a modification of the pore structure and connectivity and therefore to different LNAPL saturations.

4.4 Compare the pore size after LNAPL injection and after KI injection

Figure 6 shows retained KI in each pore for CT image after LNAPL injection and then after KI
injection, respectively. The vertical axis in each figure is ratio of pore occupied by KI in each pore size after LNAPL injection and then after KI injection. Figures 6 and 7 produce the information how much KI was occupied in each pore size after each fluid injection. As shown in Figure 6, pore sizes, which the ratio of KI occupation is less than 10%, are 85-165μm for Case1, 135-155μm for Case2, 55-95μm for Case3 and 75-105μm for Case4. From these results, LNAPL tends to choose large pore diameter which exists connecting each pore.

As shown in Figure 7, the all LNAPL completely replace KI at 195μm in Case2 and KI occupation is increased 42% at 175μm in Case3. In Case 4 also, KI occupation is increased 40% at 155μm in Figure 7. It should be also recognized that 185μm for Case 1 and 165μm for Case4 are high KI occupation rate in Figure 6. Basically, LNAPL in large pore seems to replace to KI. Probably, this conclusion can be valid in the pore space that capillary force is dominant. Due to increase of pore size, the capillary force is lost in the grain materials according to Young-Laplace’s law. In this experiment, these results are considered that not capillary pressure but hydraulic pressure more dominants to the range of grain size more than 165μm in tested materials. The pore size of 175-185μm for Case1 is corresponding to the 95-100% of maximum pore size for Case1. Similarly, the pore size discussed for Case 2, Case3 and Case 4 are corresponding to 84-100%, 90-100% and 94-100% to maximum pore size, respectively. It is presumed that hydraulic pressure is dominant in these pore size. This could be concluded because LNAPL in the largest pore replace KI as shown in Figures 6 and 7. This is a future problem that pore size shape and aspect ratio between pore spaces can affects the capillary force.

5 CONCLUSIONS

With different granular materials which are sandy soil and glass beads as a target, LNAPL and KI solution injection tests were performed and scanned by micro-focused X-ray CT scanner. Spatial distribution of pore filled with LNAPL was visualized and quantitatively evaluated by an image processing technique using marker-controlled watershed, a mathematical morphological method. Key conclusions are as follows:

1) LNAPL saturation is effected by pore shape. The acute pore tend to increase LNAPL saturation. This is because the pore shape have relation to pore connectivity.
2) System of remained LNAPL for Case1 is local. LNAPL is remained in large pore diameter.
3) System of remained LNAPL have relation to the capillary force. It is necessary to discuss how pore size area can affect the capillary force.

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