Remediation of LNAPL in sandy soil resulting from air injection

Erika Shiotai and Toshifumi Mukunoki

i) Master candidate, Department of Civil Engineering, Kumamoto University, 2-39-1, Kurokami, Kumamoto City 860-8555, Japan.  
ii) Associate Professor, X-Earth Center, Graduate School of Science and Technology, Kumamoto University, 2-39-1, Kurokami Chuouku, Kumamoto City 860-8555, Japan.

ABSTRACT

The objective of this paper is to provide results of a study on verifying the remediation of sand contaminated by LNAPL. The remediation mechanism used was air injection and a micro x-ray computed tomography (CT) scanner was used to show the effectiveness of the mechanism. First, air at 20°C and 60°C was injected into dry sandy soil. Then, the specimen was observed for effluent change. In this paper, the following three items will be shown: 1) development of the test apparatus for fluid injection into the sandy soil, 2) establishing a scan method using a micro-focused x-ray CT scanner and 3) development of a new image processing technique that can evaluate residual fluids in the pore structure of sandy soil.

Keywords: LNAPL, air sparging, X-ray CT, image analysis

1 INTRODUCTION

Air sparging method is well known as an effective remediation technique for soil contaminated by Light Non-Aqueous-Phase Liquids (LNAPL) such as gasoline and diesel fuel (David et al. 2000, Johnson et al. 1993). The process of air sparging can be defined as the injection of compressed air into the saturated zone below/within the contaminated zone. When the air is injected through the saturated zone toward the unsaturated zone, LNAPL dissolved in the ground water, sorbed onto the soil, or trapped in the soil are stripped into air channels and transferred to the unsaturated zone. However, to more accurately predict results and develop better systems, an advanced understanding of the interaction between the injection rate and capillary effect due to the remediation process is required (John and Ronald 1997, Jeffrey and Krishna 2000, and Shane et al. 2000). Several studies on visualization using a micro-focused x-ray computed tomography (CT) scanner have been conducted to better understand water and oil behavior at the pore scale. The effectiveness of using a CT device in these studies has been reported by Wildenschild et al. 2005, Culligan et al. 2006, and Mukunoki and Mikami 2013.

The objective of this paper is to provide results of a study on verifying the remediation of sand contaminated by LNAPL. The remediation mechanism used was air injection, and a micro x-ray CT scanner was used to show the effectiveness of the mechanism. First, air at 20°C and 60°C was injected into dry sandy soil. Then, the specimen was observed for effluent change. In this paper, the following three items will be shown: 1) development of the test apparatus for LNAPL injection into the sandy soil, 2) establishing a scan method using a micro-focused x-ray CT scanner and 3) development of a new image processing technique that can evaluate residual fluids in the pore structure of sandy soil.

2 METHODS

2.1 Materials

Toyura sand was used for the test as the soil material. It possesses 170 μm of effective grain size and a 1.29 uniformity coefficient. The soil density of this sand is 2.64 t/m³. The dry density of the specimen for this experiment was 1.60 t/m³, which indicates 0.393 void ratio. In this condition, the specimen possesses 5.74 ml of one pore volume.

Since the CT image is composed of a CT value that is proportional to the material density, it is possible to distinguish different phases and evaluate them separately by analyzing the CT value. In the remediated condition, the CT image will show two phases in the pore space: LNAPL and injected air. Table 1 shows the specification of tested LNAPL in this study.

| Component             | Isoparaffin |
|-----------------------|-------------|
| Density (t/m³)        | 0.75        |
| Viscosity (mPa*s)     | 1.29 (20°C) |
| Surface tension (mN/m)| 24.68       |
| Interfacial tension with water (mN/m) | 54.5 |
| Contact angle (°)     | 7.2         |

http://doi.org/10.3208/jgssp.JPN-049
2.2 One-dimensional flow injection test apparatus

The system for a one-dimensional (1D) injection experiment that can be combined with a micro focused x-ray CT scanner was newly developed to visualize the residual condition of LNAPL after air injection process. Figure 1(a) is a photograph of the experimental apparatus, and Figure 1(b) is a diagram of the entire experimental system.

(a) Photograph of 1D flow injection test apparatus
(b) Diagram of test system

As shown in Figure 1(b), the temperature of the air to be injected can be controlled by a cartridge heater, thermocouple bar, and a fan in an insulated box newly developed for this study. Hence the air temperature could be kept at 25 and 60 °C, respectively.

Each device was connected by a Teflon tube that had a 1-mm inside diameter (ID) and 1 mm thickness. The pressure sensor was installed between the syringes and the specimen column. The outlet point of the specimen column was placed above the precision balance. Also, there was a three-directional valve between the specimen column and the pressure sensor, which was used to replace one fluid with another kind of fluid. A specimen column was made of aluminum, 200 mm long and 20 mm wide, and a 10-mm inner diameter and 10 mm thickness. To prevent chemical aging caused by LNAPL, O-rings made of oil-resistant rubber (Viton) were used on the inside of the inlet and outlet points of specimen column. A wire mesh was also installed to prevent mass losses of soil particles during injection flow. The injection rate was controlled by a syringe pump—the 33 Twin Syringe pump manufactured by Harvard apparatus.

2.3 Test cases

The air injection rate causes the LNAPL displacement to be affected by the capillary effect created by the pore structure. Thus, to investigate the interaction between the air injection rate and the capillary effect, the test was conducted with two injection rates, both of which are under the effect of capillary fingering. The degree of capillary fingering was determined by the injection rate. Table 2 shows the air injection rates and displacement conditions. Both Case 1 and Case 2 are in the range of capillary fingering according to their air injection rates. As for the viscosities ratio (M), injected air was considered to be displaced with mostly water in air sparging. Hence log M was determined by the viscosity values of LNAPL and air. Once the test was started, the injection fluid was pushed out from the syringes. Then, it passed through the pressure sensor and infiltrated the specimen column from the inlet point. When the fluid came out of the outlet point of the specimen column, it dropped into the precision balance. Thus, this system let us follow the temporal changes of the injection pressure and the discharged fluid mass during the experiment.

Table 2. Test cases

| Injection rate | Air temperature | log M |
|----------------|-----------------|------|
| Case 1         | 50 (ml/h)       | 25 (°C) | -1.74 |
| Case 2         | 50 (ml/h)       | 60 (°C) | - |

2.4 Scan condition

Table 3 shows the scan condition of micro x-ray CT in the X-Earth Center at Kumamoto University. The power voltage and current of scanner were 180kV and 200 μA, respectively, and these values are relatively high. The reason why these values were used is that the sandy soil was installed in an aluminum cylinder. Hence, it was necessary to generate an x-ray beam strong enough to penetrate the specimen, which required a larger voltage.

The resolution (i.e. the minimum pixel dimension) is 6 μm with a 14.2 cm focus-center distance (FCD). A flat panel detector (FPD) can perform 3D scanning using a cone beam x-ray.

Table 3. Scan condition.

| Voltage (kV) | Current (μA) | Number of view angles | Projection average | FCD (mm) | Resolution |
|-------------|-------------|-----------------------|-------------------|----------|-----------|
| 180         | 200         | 1500                  | 10                | 14.2     | 6         |

2.5 Test procedure

The test procedure consists of three steps as follows: Step 1 (Creation of contaminated sand) A test LNAPL of 2 pore volume (PV) is injected into the completely dry sand at a flow rate of 25 cm³/h and then the specimen was scanned.
Step 2 (Remediation with air injection)
Air of 5 PV at each temperature (25°C and 60°C) was injected at a flow rate of 50 cm³/h. Simultaneously the mass of effluent and air pressure were measured. After the injection was finished, the specimen was scanned.

Step 3 (Remediation by evaporation with air injection)
Air of 5 more PV at each temperature (25°C and 60°C) was injected at a flow rate of 50 cm³/h. Simultaneously the mass of effluent and air pressure were measured. After the injection was finished, the specimen was scanned.

3 RESULTS AND DISCUSSION

3.1 Pressure profile due to air injection
Figure 2 shows the pressure profile due to injecting air with 25°C and 60°C as Case 1 and Case 2, respectively. In Step 2, pressures for Case 1 and Case 2 increased immediately and significantly just after the start of air injection. Eventually, the pressure for Case 1 reached 4.1 kPa, but the pressure for Case 2 did not. The steady state pressure for Case 1 and Case 2 were 1.34 kPa and 1.23 kPa, respectively. Injecting air at high temperature reduced the viscosity of the LNAPL; hence, the pore pressure of LNAPL for Case 1 was greater than Case 2.

![Pressure profile due to air injection](image)

In Step 3, the maximum pressure for both Case 1 and Case 2 was 2.17 kPa—less than in Step 2. The chief air flow path was created previously in Step 2. The steady state pressure in Step 3 was one-third of the maximum pressure in Step 2. Interestingly, the entry pressure was still detected in Step 3. If the air flow path was created completely in Step 2, the Step 3 entry pressure should not be measurable. However, the pressure profile in Step 2 showed that the pressure condition reached a steady state in the specimen.

3.2 Discharged-mass profile due to air injection
Figure 3 shows the mass of LNAPL recovered from the specimen at the effluent position in Step 2. As for Case 1, it took 300 seconds to measure the mass of LNAPL; however, it took 353 seconds for Case 2. These results indicate that air flow at 60°C reduced the viscosity of LNAPL such that in Case 2 more LNAPL came out than in Case 1. At the end of Step 2, the recovered mass of LNAPL for Case 1 and Case 2 was 0.98 g and 1.41 g, respectively.

In addition, pore volume can be evaluated by the actual inlet itself and it was 5.74 ml. Therefore, the saturation degree of LNAPL can be evaluated. LNAPL saturation was reduced at Step 2 by 22.7% for Case 1 and 26.5% for Case 2. In fact, there were no mass of LNAPL in Step 3. Probably, remained LNAPL in Step 2 would be trapped LNAPL and they should be removed by evaporation process.

![Discharged mass profile due to air injection](image)

3.3 Proof of LNAPL extraction using image subtraction and multiplication
Figure 4 shows an X-ray CT image before air injecting. As shown in Figure 4, three kinds of materials can be observed: sand particles, LNAPL, and air. After air injection, three behaviors are identified as follows: A) Retained LNAPL, B) Moved LNAPL, and C) Trapped LNAPL.

A) Retained LNAPL in initial position;
B) LNAPL moved from initial position by injected air
C) Trapped LNAPL by pore space in which KI solution existed in initial condition.

3.4 Image segmentation
As previously mentioned, the obtained CT images show three phases such as soil particles, air, and LNAPL. However, the CT value histogram of the images was not suitable for using the Otsu method (Otsu 1979). Therefore, the marker-controlled watershed method, which does not lose the spatial geometrical information from the CT value, was applied to the images. The algorithm can be referred from Mukunoki and Mikami (2013).
3.5 Image multiplication and subtraction

Image multiplication was applied to extract LNAPL blobs on the basis of the displacement conditions. Figure 5 presents binary images as the process of calculation. In the figures, black voxels are shown with a binary value of 1 and white voxels are shown with that of 0. Figure 5 (a) is presented as LNAPL blobs and Figure 5 (b) is presented as injected air. They are considered to be extracted from the same cross-sectional CT image of the same specimen. When Figure 5 (a) is multiplied by (b), four combinations of voxel values result from the images as shown in Table 4. Among them, only the combination of (4) can retain the binary value for the case of LNAPL blobs and injected air located in the same voxel coordinate. Figure 5 (c) presents the result of the multiplication. Any black voxels in Figure 5 (c) are considered as LNAPL blobs that are retained with LNAPL at Step 1. The Moreover, image subtraction makes it possible to evaluate the black area of Figure 5 (d) indicating moved LNAPL and the black area of Figure 5 (e) indicating retrapped LNAPL. In this way, it is possible to extract LNAPL blobs on the basis of displacement conditions resulting from injected air.

Table 4. Image multiplications.

| Pattern | Voxel value of (a) | Voxel value of (b) |
|---------|-------------------|-------------------|
| (1)     | 1                 | x                 |
| (2)     | 0                 | x                 |
| (3)     | 0                 | x                 |
| (4)     | 1                 | x                 |

3.6 Volume ratio of LNAPL with each status

Figure 6 shows the x-ray CT images of each stage of LNAPL (retained, moved, and trapped) in one 2D image for Case 1. Because the x-ray CT image is a digital image it provides a great advantage for quantitative evaluation during image analysis. This image processing was done 3-dimensionally so we could evaluate the LNAPL volume for each status. Figure 7 shows the pie graph of LNAPL obtained from measuring the voxel of LNAPL at each stage. The ratio of moved LNAPL was 43% for Case 1 and 56% for Case 2. Meanwhile, retrapped LNAPL was 16% for Case 1 and 37% for Case 2. These results indicate that moved LNAPL also produced retrapped LNAPL. A comparison of the saturation of LNAPL in Case 1 with that of Case 2, showed a difference of only 4%. However, image subtraction and multiplication proved that the LNAPL behavior was different in the two cases. The greater amount of moveable LNAPL in Case 2 caused some LNAPL to be trapped due to air injecting.
**Figure 6.** The x-ray CT images showing each stage of LNAPL extraction: retained, moved, and trapped.

**3.7 Size of pore space filled with LNAPL**

Figure 8 shows histogram charts with respect to pore size in the sample obtained from the x-ray CT image analysis developed by Mukunoki and Mikami 2013. The value of the vertical axis is the ratio of each pore size normalized with the total number of pore sizes at Step 1 and Step 2. Hence, they show the saturation of each stage of LNAPL. Retained LNAPL was mostly distributed in pores of size 42 μm. The maximum ratio of LANPL saturation degree for retained LNAPL was 17.2 % for Case 1 and 10.4 % for Case 2, respectively. The maximum diameter of pore size with LNAPL as a blob was 10.2 μm for Case 1 and 78 μm for Case 2. The diameter of moved LNAPL for Case 2 was between 6 μm and 54 μm and the ratio was 83.5 %. Meanwhile, it was 60.5 % for Case 1. This means the specimen in Case 2 had smaller blobs than Case 1. In retrapped LNAPL, the diameter for both cases was less than 54 μm and its saturation was 90%. This indicates that the dimension of retrapped LNAPL was quantitatively smaller than that of moved LNAPL.

**4 CONCLUSIONS**

In this study, sandy soil contaminated by LNAPL was scanned by a micro-focused x-ray CT scanner and the fluid behavior and evaporation properties of LNAPL were evaluated using visualization and image analysis. Some conclusions are:

1) The mass recovered from the specimen with 60°C (Case 2) air injection was greater than that with 20°C (Case 1) and the entry pressure for Case 2 was less than Case 1 because the higher temperature caused the fluid to be less viscous.

2) Injecting air at 60°C caused movement of the LNAPL.

3) When injecting air at 60°C, the LNAPL blobs with smaller than 54 μm remained as retrapped LANPL.

4) Retrapped LNAPL means non-movable LNAPL; hence, evaporation would be expected as the next step of the process of physical remediation.

**ACKNOWLEDGEMENTS**

This research was financially supported by a Grant-in-Aid for Scientific Research (C) No. 30423651. The authors thank Mr. Kazuaki Mikami (JOGMEC) and Mr. Kenichi Sugimura and Mr. Naoki Tsukamoto, graduates of the graduate school of science and technology, Kumamoto University.
Figure 8. Distributions of pore size of each type of LNAPL blob

REFERENCES

1) Culligan, K.A., Wildenschild, D., Christensen, B.S.B., Gray, W.G and Rivers, M.L. (2006): Pore-scale characteristics of multiphase flow in porous media: A comparison of air-water and oil-water experiments, Advances in Water Resource, Vol.29, pp.227-238.

2) David H., Beckett, G.D. : Persistence of LNAPL sources: relationship between risk reduction and LNAPL recovery, Journal of Contaminant Hydrology, Vol.59, pp.3-26, 2002.

3) Jeffrey, A.A. and Krishna, R.R. (2000): Removal of dissolved- and free-phase Benzene pools from ground water using in situ air sparging, Journal of Environmental Engineering, Vol.126, No.8, pp.697-707.

4) Johnson, R.L., Johnson, P.C. and McWhorter D.B. (1993) : An Overview of In Situ Air Sparging, Ground Water Monitoring & Remediation, Vol.13, Issue.4, pp.127-135.

5) John, E. M., Ronald, W. E.(1997) : Numerical simulation of air sparging for remediation of NAPL contamination, Ground Water, Vol.35, No.1, 99-110.

6) Mukunoki, T. and Mikami, K. (2013), Study on mechanism of two-phase flow in porous media using X-ray CT Image Analysis, Proc. of the 18th International Conference of Soil Mechanics and Geotechnical Engineering, TC106 selected paper, pp.1163-1166.

7) Otsu N. : A Threshold Selection Method from Gray-Level Histograms, IEEE Transactions of systems, man, and cybernetics,Vol.9(1), pp.62-66, 1979.

8) Shane, W.R., Say, K.O. (2000): Influence of porous media, airflow rate, and air channel spacing on benzene NAPL sparging removal during air, Environ. Sci. Technol, Vol.34, pp.764-770.

9) Wildenschild, D., Hopmans, J.W., Rivers, M.L. and Kent, A.J.R. (2005): Quantitative analysis of flow processes in a sand using synchrotron-based x-ray microtomography, Vadose Zone Journal, Vol.4, pp. 112-126.