

Migration of hydrocarbon liquid in unsaturated laterite soil with dissimilar volume and moisture content

M F Abd Rashid¹, N Alias¹, K Ahmad², R Sa'ari¹, M Z Ramli³, Z Ibrahim¹ and M I Shahrin²

¹Department of Water and Environmental Engineering, School of Civil Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Johor, Malaysia.

²Department of Geotechnics and Transportation, School of Civil Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Johor, Malaysia.

³Institute of Noise and Vibration, School of Civil Engineering, Faculty of Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Johor, Malaysia.

Email: fadhli@civil.my

Abstract. Groundwater contaminations due to hydrocarbon or organic solvent spills in saturated or unsaturated zones affect the water resources. The quantity of hydrocarbon spills in the groundwater might influence the hydrocarbon migration. In addition, the soil moisture content in influencing the rate of migration is also unknown. Therefore, this paper presents the investigation migration of Light Non-Aqueous Phase Liquid (LNAPL) in unsaturated laterite soil with two dissimilar moisture contents and different of LNAPL volume. These lab-scale studies discuss the results of migration phenomena by employing image analysis by using soil column, mirror, LNAPL, and Nikon D90 digital camera. Aggregated soil was poured and compressed in an acrylic column until 100 mm height. Then LNAPL was poured onto the soil column surface instantaneously. The LNAPL migration pattern in laterite soil was monitored and recorded using digital image processing technique (DIPT) at certain time intervals then were processed with Surfer software and Matlab routine for plotting the LNAPL migration pattern. As a result, the analysis displayed the higher rate migration of LNAPL with high moisture content and bigger amount of LNAPL volume. However, the rates of LNAPL migration decrease with lower moisture content and small amount of LNAPL volume. The migration time required to reach bottom of soil was longer for low moisture content with smaller amount of LNAPL volume as compared to the high moisture content with bigger amount of LNAPL volume.

1. Introduction

An important role is held by groundwater as it assists the development the world's potential clean water resource. Thus, the increasing threat of subsurface contamination posed towards groundwater must be avoided [1]. An establishment of integrated water resource management, as well as urban planning, maintainable cities, catastrophe preparation, initial systems of warnings have been made across Asia [2]. Climate change has been found to be a factor that affects the water sector [3]. The quality and quantity of water availability at a global and local scale are mostly effected by a country's rising water demand that come from the changes in the industrial and agricultural industries [3-5]. The environmental issue of soil contamination and groundwater caused by non-aqueous phase liquids (NAPLs) has been found to become a largely alarming problem [6-7]. According to [8] upon releasing



LNAPL on a soils' subsurface, the LNAPL will penetrate into the soil then floating on the water table groundwater. Whereas after releasing Dense Non-Aqueous Phase Liquid (DNAPL) to the subsurface soil, the DNAPL will migrate to the ground until it reaches the saturated zone [9] because to its density is more dense than groundwater. [10] had stated that three phases are able to exist in the aqueous phase, non-aqueous phase and in gaseous phase and they are able to contain organic contaminants.

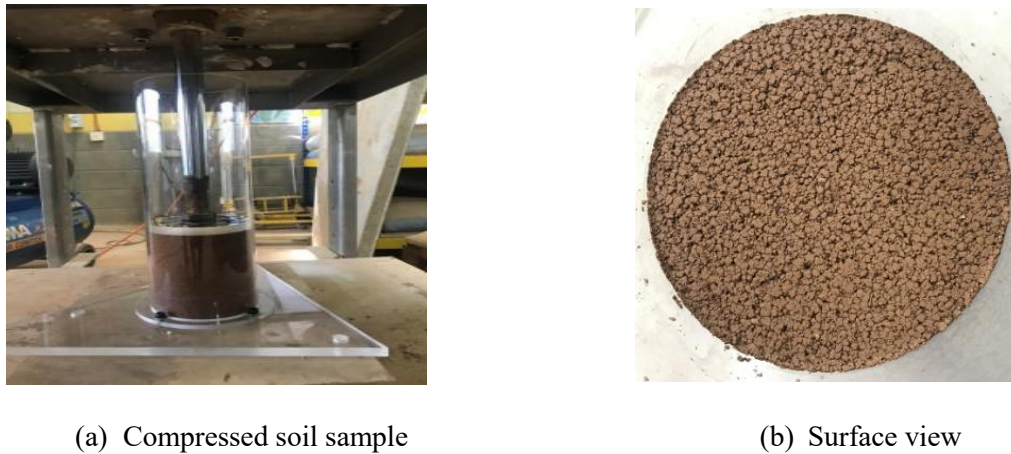
Double porosity media is defined as a soil with two particular porosity media scales [11]. [12] had stated that the characteristics of double-porosity media can be seen from the difference in hydraulic properties and size of the pores with two definite sub-regions in soil. In the study carried out by [13 – 14] the inter-aggregated and intra-aggregated in the soil confirmed the porosity. According to [15 – 16] compacted soils and agricultural top-soils are example of soils that display hydraulic properties and pore-size bimodal distribution that have inter and intra-aggregate pores for aggregated innate soil. According to [17], groundwater pollution is caused by the unwanted changes of groundwater quality that stems from the influence triggered by human activities. [13 – 14] stated that an influential factor of fluid movement in the subsurface system is expected to be the high value of permeability. [18] had found that the type of contaminants that are produced and the degree of is localization as well as its' loading history are the characteristics of groundwater pollution. [19] has called for an immediate urgency to assess the zones of contamination and upon doing so there should be an effort to design schemes of remediation. Double-porosity soil characteristics can be re-created by using infiltration tests that are one-dimensional for experiments [20]. [21] had stated that in order to ensure non-intrusive and non-destructive methods are used; a lot of measuring tools are required to monitor and record liquid migration. A higher precision of observation and system characterization can be obtained from experiments that involve non-invasive imaging techniques [22]. Of late, more researchers have contributed findings focusing on double-porosity soil where experiments are made in physical laboratories. These studies are from [12, 19, 31 – 34, 23 – 30]. These findings have contributed to a further understanding of double-porosity soil and its' characteristics. According to [1], to minimize the effects from existing or proposed activities in the agricultural, municipal or industrial sectors towards the quality of groundwater, experiments on contaminants are necessary.

Therefore, to analyse the penetration of liquid in soil media using digital image processing technique (DIPT) the contaminant migration through laterite soil must be studied in order to close the research gap. Two (2) objectives were recognized and set as a basis for this study which are; (i) to examine inter and intra-aggregate on the soil properties of double-porosity, and (ii) to define the pattern of migration of red dyed-oil in aggregated laterite soil by exerting different moisture content and volume of LNAPL of the surface of the soil and analysing using digital imaging process techniques. Section 2 displays preparation of soil as well as the penetration of LNAPL through the soil as captured using the digital image processing technique. The hue-saturation-intensity plot which explains the image analysis is portrayed in Section 3 and also presents the discussions and result findings. At the end of the paper, Section 4 gives the conclusion of this study.

2. Materials and physical modelling methodology

This section briefly explains the laboratory procedures and laboratory scale setup which encompass the soil samples, apparatus and aggregation. The samples for soil were taken from the School of Electrical Engineering, Faculty of Engineering in Universiti Teknologi Malaysia, Johor and were then prepped into multi-phase soil with reference [35] and conduct laboratory soil properties based on the [36]; consisting of; liquid limit, plastic limit, density of particles and classified based on the [37] Unified Soil Classification System (USCS). The samples were carried out using different water contents of 25% and 30%. The laterite soil was mixed with a water content of 25% and 30% to obtain the samples which were labelled as Sample 1 (a), Sample 1 (b), Sample 2 (a), and Sample 2 (b). All the samples were kept for a minimum of 24 hours in a cool temperature to enable curing and to avoid evaporation re-sealable plastic bags were used. All samples were sieved through 2.36 mm siever to form an aggregated soil. Then placed and compressed in an acrylic column using a compression machine with 1kgf/cm^a until the height reached 100 mm. in order to inspect and observe the LNAPL migration, apparatus made of acrylic material was used where the original soil sample setup was not

destroyed or intruded at the circumference surface. The position of the soil columns, arrangement of acrylic soil column and the V shaped mirror was setup for each soil sample as shown in Figure 1.

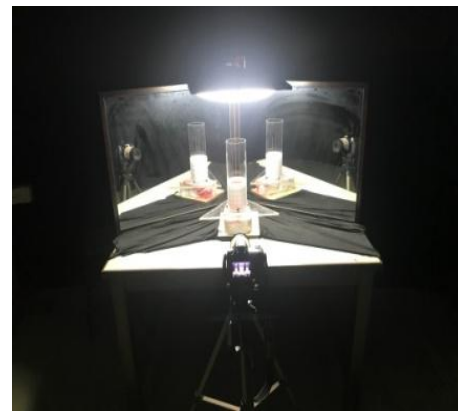


(a) Compressed soil sample

(b) Surface view

Figure 1. Soil sample preparation.

The dimensions of the acrylic sample used for the experiments were of 300 x 94 mm, height x inner diameter with a sealed base design. This design enabled a better visualization to observe the all area of acrylic sample. Figure 2 and Figure 3 display the arrangement of the experimental procedure. The image acquisition system for migration of hydrocarbon liquid was captured using a D90 DSLR Nikon camera that was fixed with 23.6 X 15.8 mm a sensor size and 2136 X 3216 pixels image format, where 5.6 X 5.6 μm sizes were set for each pixel. For all the experiments, the ISO speed and digital camera aperture setup were set at 2500 and f/5.6 respectively. The samples were sheathed with pre-drawn gridline as a control point on the reference image (of a size set at 20 mm x 20 mm) on white paper and put onto the soil column.

**Figure 2.** Whole acrylic column.**Figure 3.** Digital image experimental setup.

The experiments began by instantly pouring the red dyed toluene at the center of the surface of the aggregated laterite soil in the acrylic soil column. In order to snap the migration of the red dyed toluene, images were taken at specific time intervals. The computer software that was used to carry out the image processing was Surfer Software and Matlab routine. Images captured were saved in JPEG format and were transferred to the computer. Control points on reference image were digitalized to extract the coordinates. The boundary area of migration refer to the area of control points image for all the samples, namely Sample 1 (a), Sample 1 (b), Sample 2 (a), and Sample 2 (b) and is as shown in Figure 4.

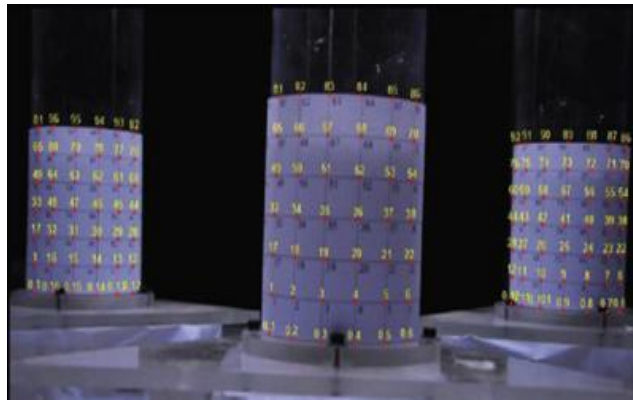


Figure 4. Control point for each references.

3. Results and discussion

Based on Unified Soil Soil Classifications System (USCS) the soil sample used was classified as Clay with High Plasticity (CH). Table 1 displays the detail of the laterite soil samples.

Table 1. Laterite soil properties

Property	Unit	Value
Liquid Limit	(%)	66
Plastic Limit	(%)	33
Plasticity Index	(%)	33
Particle Density	(Mg/m ³)	2.74
USCS	Classification	(CH) Clay with High Plasticity
Permeability, K _{average}	(m/s)	4.62 X 10 ⁻⁷

The double-porosity at the magnification folds of 180, 1000 and 3000, with moisture content of 25% and 30% is displayed in Figure 5. The SEM test shows the granular form of soil sample at 180-fold magnification. On the other hand, the inter-aggregate pores are seen at 1000-fold magnification. When the magnification of SEM test at 180-fold was done, it was found that the inter-aggregate pores had split up among themselves. Upon further magnification at 3000-fold, indication of intra-aggregate pores were identified. From these observations, it was confirmed that the formation of double-porosity laterite soil was generated by the deformable characteristics of intra and inter-aggregate pores. All samples had displayed a similarity where the soil was found to be coated by a layer of liquid when viewed using the SEM image zoom in that gave off a shining image. It was found, having coarse granules that were displayed in the moisture soil samples. Thus, these condition are expected lead to the speed and migration of liquid seeping through the soil sample.

Figure 6 and Figure 7 show the downward migration pattern in the aggregated double-porosity laterite soil sample based on the 25% moisture content for Sample 1 (a) and Sample 1 (b), additionally Sample 2 (a) and Sample 2 (b) display the 30% moisture content. The liquid penetration was one-dimensional in the soil column; meanwhile, when the curve joint of the right and left boundaries formed in the HSI plot merge it can be clearly seen as two-dimensional. All images were captured with certain intervals time as shown in Table 2 [32], [33].

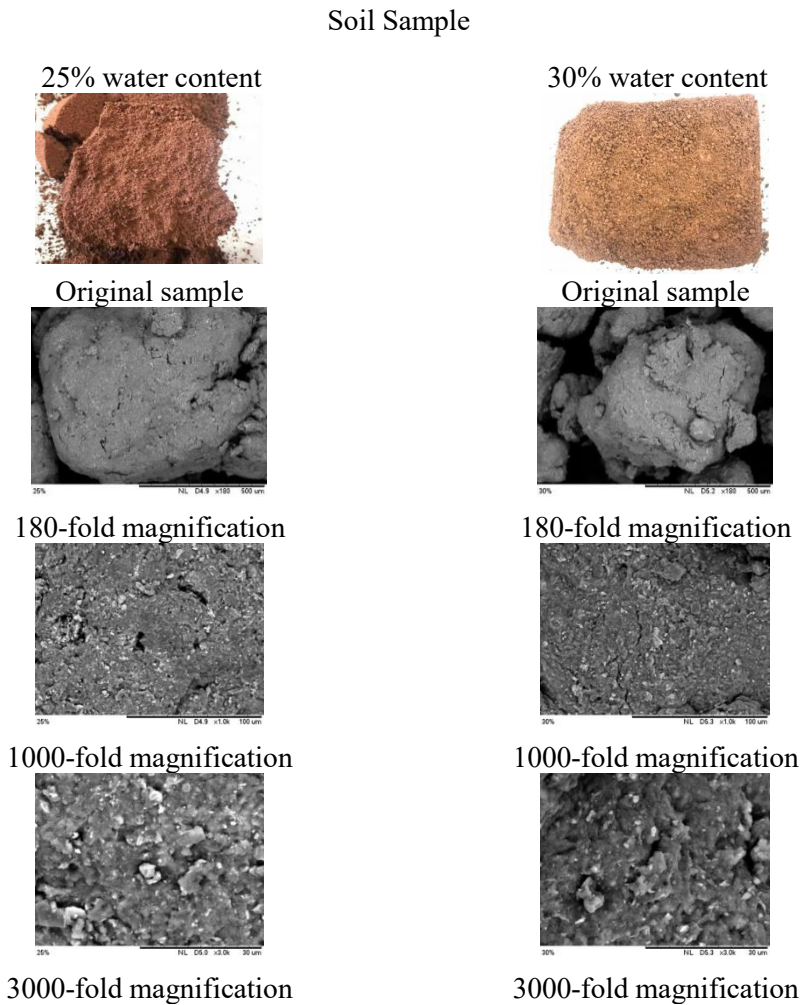


Figure 5. SEM magnification image for 180, 1000, and 3000-fold.

Table 2. Frequency of image acquisition.

Period	Frequency Interval
0 – 1 st minute	5 seconds
1 st minute – 2 nd minute	10 seconds
2 nd minute – 3 rd minute	20 seconds
3 rd minute – 4 th minute	30 seconds
4 th minute – 5 th minute	30 seconds
5 th minute – 6 th minute	1 minute
6 th minute – 7 th minute	1 minute

As observed in Figure 6 and Figure 7, the red dyed-oil toluene migrated downward and moved in one-direction due to it being poured instantly using a glass funnel at the centre of the soil sample surface. It was found that for these samples, before the whole sample surface was covered, the hydrocarbon liquid would have migrated into the laterite soil sample beforehand.

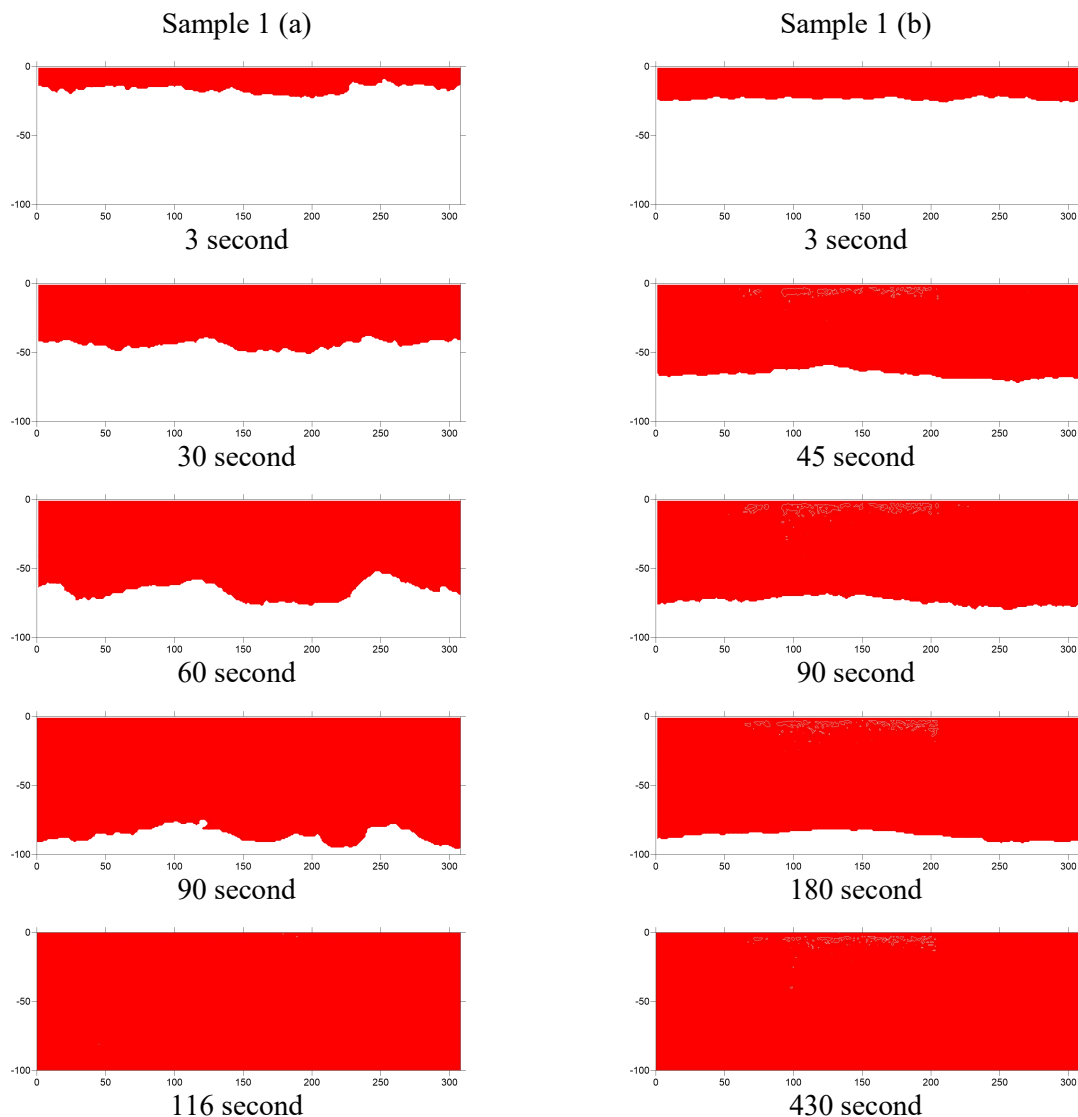


Figure 6. HSI plots on aggregated laterite soil with 25% moisture content.

Sample 1 (a) recorded HSI data of hydrocarbon liquid penetration at intervals of 3, 30, 60, 90, and 116 seconds, Sample 1 (b) at intervals of 3, 45, 90, 180, and 430 seconds, Sample 2 (a) at intervals of 3, 15, 30, 45, and 72 seconds, and Sample 2 (b) at the intervals of 3, 30, 60, 90, and 132 seconds as can be seen in Figure 6 and Figure 7. Based on the results, the intervals for each image show same depth of migration with different durations. According to the observation of the contour plot results on HSI intensity in Sample 1 (a), Sample 1 (b), Sample 2 (a) and Sample 2 (b), it was found that the migration pattern flow was moving downward consistently of horizontal line at x-axis due to multi-phase character soil. At 3 seconds after the instantaneous pouring, all samples displayed that the migration of the red dyed toluene had reached a quartile height of the soil sample. At the initial stage, the migration rate was average for all samples. However, the second HSI image demonstrates that the rate of migration is affected by the content of moisture and the amount of LNAPL that is exerted on soil surface. Therefore it can be concluded that the soil sample of Sample 1 (a) and 1 (b) had the slowest migration rate when compared to Sample 2 (a) and 2 (b) as it shows that the bigger the volume of LNAPL, the faster the migration rate. Furthermore, it was observed that the time taken for the red dyed oil to reach to the bottom of the soil samples that had a lower moisture content and smaller

volume of LNAPL had the slowest migration in comparison to the soil samples with higher moisture content and larger amount of LNAPL exerted on the soil surface.

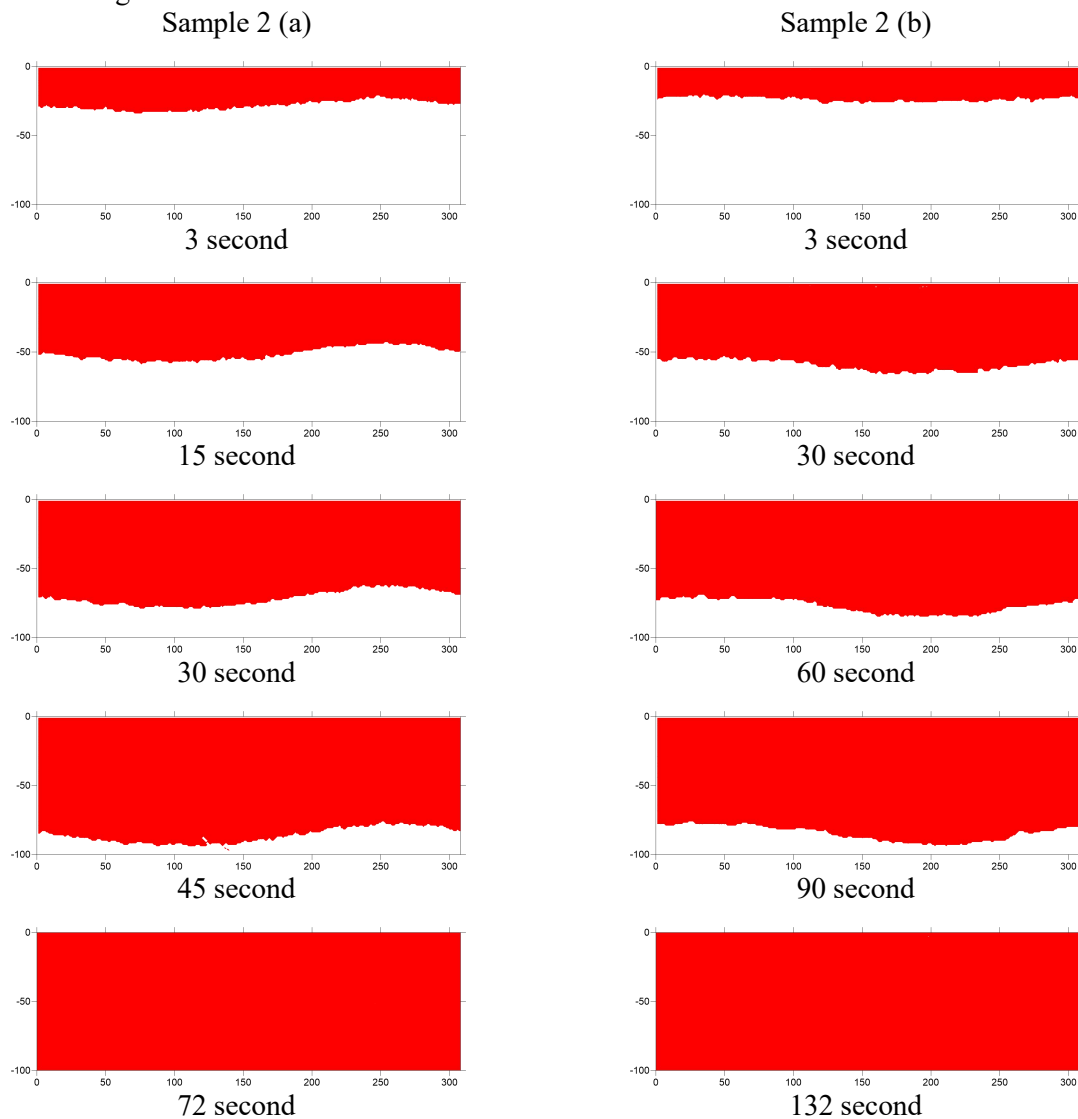
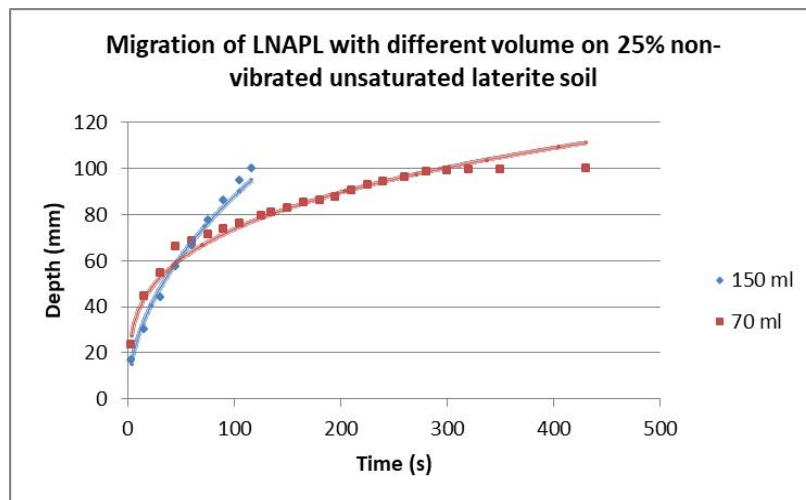
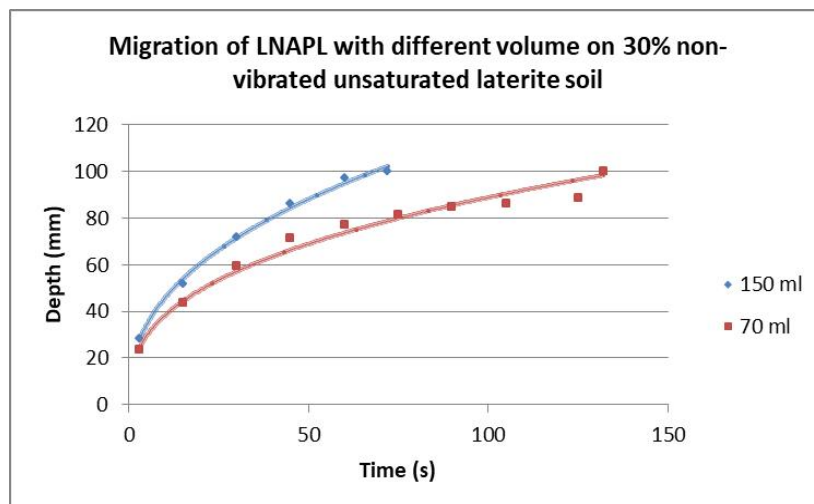


Figure 7. HSI plots on aggregated laterite soil with 30% moisture content

As shown in Figure 8, the rate of migration of soil Sample 1 (a) and 1 (b) is 1.69 mm/s and 1.25 mm/s, which was slower than soil Sample 2 (a) and 2(b) which is 3.37 mm/s and 2.00 mm/s. Due to the immiscible hydrocarbon liquid interaction with the presence of low moisture content of 25% compared to soil Sample 2 (a) and soil Sample 2 (b) at 30%, the liquid migration time was comparably longer for Sample 1 (a) and Sample 1 (b). This occurred because the air voids in the soil were filled with water, thus increasing the migration rate in the soil Samples of 1 (a) and soil Sample 1 (b) compared to the soil Samples 2 (a) and soil Sample 2 (b). Another factor that affects the migration time is the volume of LNAPL amount that is exerted on the soil surface as it increases the pressure due to the oil height on the soil surface, filling up the pores between the inter-aggregate and intra-aggregates that act as double-porosity. Soil Sample 1 (a) and soil Sample 2 (a) demonstrate that the migration rate was faster compared to the soil Sample 1 (b) and soil Sample 2 (b).



(a) 25% moisture content with different LNAPL volume.



(b) 30% moisture content with different LNAPL volume.

Figure 8. LNAPL migration graph varies with different moisture content and different volume of LNAPL amount

4. Conclusion

Laboratory scale were conducted had shown the results of migration rates and behaviour patterns on multi-phase unsaturated laterite soil with different amounts of LNAPL volumes and distinct water contents. In order to observe the migration behaviour and pattern of liquid, red dyed-oil was poured through aggregate laterite soil that was placed in a soil column model. A test were verified and confirmed that the deformable multi-phase unsaturated soil had intra-aggregate and inter-aggregated using Scan Electron Microscopy (SEM). To analyse and extract HSI data from the digital images, techniques of digital processing was applied using the software Surfer (version 10) and Matlab routine. Upon comparison of all four (4) soil samples tested in this study, the fastest migration rate was displayed in Sample 2 (a) due to the volume pressure placed on the surface of the soil as well as the moisture content changes. The slowest migration rate was shown in Sample 1 (b) as it had a lower content of moisture and a smaller amount of LNAPL that was placed on the soil surface. It has been discovered that the soils are problematic double-porosity soil since characteristics of multi-porosity have been found from the soil samples. This study has confirmed that migration speed rate is affected by different water contents in double-porosity soils and the smaller the amount of LNAPL that is

present the slower the migration. High permeability value has also been found to influence the fluid movement on the subsurface system. From the readings given by the HSI value contour plot of the migration behaviour of red dyed-oil, future and current researchers shall be able to gain a better understanding and explain on the details specifically with the use of close range photogrammetry technique using digital camera images. This study will help to identify the behavior of actual soil condition in Malaysia (majority laterite soil) on gas station storage tank either with small or bigger volume spillage with raining and dry season in Malaysia. As a conclusion, it may help the government to create a new or improve standard operation procedure for remediation, precaution and cost for treatment on pollution of groundwater that totally useful for covering up the water shortage from water surface.

Acknowledgments

This research study was supported under the Research University Grant-GUP Tier 1 vote number Q.J130000.2522.20H53 and Fundamental Research Grant from Ministry of Higher Education Malaysia (MOHE) vote number RJ130000.7822.4F894. The authors of this study would like to thank their respective Universities, the Engineering Seismology and Earthquake Engineering Research Group (eSEER), Survey Unit, Hydraulic and Hydrology Laboratory and Geotechnical Laboratory School of Civil, Faculty of Engineering, Universiti Teknologi Malaysia for the cooperation received in the process of this research. Appreciation is also expressed towards the Kings' Scholarship and UTMLLead for their support and for all those who have been involved either directly or indirectly in this study.

References

- [1] Freeze R A and Cherry J A 1979 *Groundwater*
- [2] Sarkodie S A and Strezov V 2019 *Sci. Total Environ.* **656** 150–164
- [3] IPCC 2016 IPCC AR5 Part A WG II Final
- [4] United Nations 2015 Resolution A. Transform our world 2030 agenda Sustain. Dev. **16301** 1–35
- [5] UNDP, WWDR 2015: Water for a sustainable world.
- [6] Fagerlund F and Niemi A 2007 *J. Contam. Hydrol* **89** (3–4) 174–198
- [7] Luciano A Viotti P and Papini M P 2010 *J. Hazard. Mater* **176** (1–3) 1006–1017
- [8] Mercer J W and Cohen R M 1990 *J. Contam. Hydrol* **6** (2) 107–163
- [9] Kong N S, Abd. Rahman N, Ahmad K and Lewis R W 2012 *Sci. Res. Essays* **7** (38) 3243–3250
- [10] Environment Agency 2018 Environment Agency 1–56
- [11] Carminati A, Kaestner A, Lehmann P and Flühler H 2008 *Adv. Water Resour.* **31** (9) 1221–1232
- [12] Ngien S K, Rahman N A, Bob M M Ahmad K Sa'ari R and Lewis R W 2012 *Transp. Porous Media* **92**(1) 83–100
- [13] Mohd Fadhli A R Kamarudin A Noraliani A Zulkiflee I Mohd Zamri R and Radzuan S 2019 *MATEC Web Conf.* **255** 02007
- [14] Abd Rashid M F Alias N Ahmad K Zamri Ramli M and Ibrahim Z 2019 *IOP Conf. Ser. Earth Environ. Sci.* **220** 012037
- [15] Li X and Zhang L M 2009 *Can. Geotech. J.* **46** (2) 129–141
- [16] El-Zein A, Carter J P and Airey D W 2006 *Int. J. Numer. Anal. Methods Geomech.* **30** (7) 577–597
- [17] Harter T 2003 *Agric. Nat. Resour.* **8084** (11.2) 1–6
- [18] Domenico P A and Schwartz F W 1998 *Physical and chemical hydrogeology, Volume 1*
- [19] Zheng F Gao Y Sun Y Shi X Xu H and Wu J 2015 *Hydrogeol. J.* **23** (8) 1703–1718
- [20] Lewandowska J, Szymkiewicz A, Gorczewska W and Vauclin M 2005 *Water Resour. Res.* **41** (2) 1–14
- [21] Werth C J, Zhang C, Brusseau M L, Oostrom M and Baumann T 2010 *J. Contam. Hydrol.* **113**(1–4) 1–24

- [22] Agaoglu B Coptly N K Scheytt T and Hinkelmann R 2015 *Adv. Water Resour.* **79** 162–194
- [23] Konyukhov A and Pankratov L 2016 *Comptes Rendus - Mec.* **344**(7) 510–520
- [24] Darnault C J G, Throop J A, Dicarolo D A, Rimmer A, Steenhuis T S and Parlange J 1998 *Y. J. Contam. Hydrol.* **31**(3–4) 337–348
- [25] Kechavarzi C Soga K and Wiart P 2000 *J. Contam. Hydrol.* **46**(3–4) 265–293
- [26] Kechavarzi C Soga K and Illangasekare T H, 2005 *J. Contam. Hydrol.* **76**(3–4) 211–233
- [27] Alazaiza M Y D Ngien S K Bob M M Kamaruddin S A and Ishak W M F 2017 *Transp. Porous Media* **117**(1) 103–123
- [28] Alazaiza M Y D Ngien S K Bob M M Kamaruddin S A and Faizal Ishak W M 2017 *Int. J. Geotech. Eng.* **11**(3) 316–320
- [29] Alaziya M Y D Ngien S K, Bob M M Kamaruddin S A and Faizal Ishak W M 2016 *MATEC Web Conf.* **47** 03023
- [30] Kamaruddin S A Sulaiman W N A Zakaria M P Othman R and Rahman N A 2011 *Natl. Postgrad. Conf.* 1–7
- [31] Foong L K Abd Rahman N and Ramli M Z 2016 *Malaysian Journal of Civil Engineering* **28**(3) 207-222
- [32] Foong L K, Abd Rahman N, Nazir R and Lewis R W 2018 *Geol. Croat.* **71** (2) 55–63
- [33] Sa'ari R Abdul Latiff N H Abd. Rahman N Mohamed Yusof Z Mustaffar M Hezmi M A Ngien S K and Kamaruddin S A 2015 *J. Teknol.* **3** (72) 23–29
- [34] Ngoc T D T, Lewandowska J and Bertin H 2007 *18ème Congrès Français Mécanique Août 2007* January 27–31
- [35] Bagherieh A R, Khalili N Habibagahi G and Ghahramani A 2009 *Eng. Geol.* 105(1–2) 44–50
- [36] BS 1377-2:1990 1990, *Methods of test for Soils for civil engineering pueposes - Part 2: Classification tests* (London)
- [37] American National Standards Institute 2019, *D2487-17 Standard Practice for Classification of Soils for Engineering Purposes (Unified Soil Classification System)* in *ASTM D2487 - 17* (West Conshohocken) pp 10