UNIVERSITI PUTRA MALAYSIA

ASSESSMENT OF THE DISTRIBUTION PROFILE OF LIGHT NONAQUEOUS PHASE LIQUID IN UNSATURATED ZONE UNDER THE INFLUENCE OF RAINFALL RECHARGE

SAMIRA ALBATI BINTI KAMARUDDIN

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By

SAMIRA ALBATI BINTI KAMARUDDIN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

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Chair: **Associate Professor Wan Nor Azmin bin Sulaiman, PhD**

Faculty: **Environmental Studies**

Leaking from underground storage and surface spills of hydrocarbon sources can cause serious light non-aqueous phase liquid (LNAPL) contamination in subsurface environments. In real conditions, the multiphase flow during LNAPL migration can be affected by rainfall recharge. To consider this, a study was carried out to investigate the distribution of LNAPL migration in the unsaturated zone through qualitative and quantitative experiments, as well as numerical simulations. Both qualitative and quantitative experiments utilized light reflection method (LRM) for NAPL saturation imaging technique.

In the qualitative experiment, the image analysis used conventional calibration relationship to determine the distribution profile of LNAPL in a 2-D model. The penetration depth of benzene and toluene after 24 h of injection initiated was 37.0 cm and 33.4 cm, respectively. At the same time, the toluene plume occupied larger area (30.7%) compared to benzene plume (25.2%). The benzene moved deeper as

expected due to the lower retardation factor, R and higher water solubility compared to toluene. More benzene was volatilized because its vapor pressure is higher than toluene. The differences showed that the chemical properties of the LNAPL source have considerable influence on their transport mechanism through porous media. Rainfall recharge showed minimal effects to benzene and toluene distribution due to its volatilization mechanism in porous media. In the quantitative experiment, multispectral imaging technique was applied to develop reliable image analysis. The average optical density (OD) from the captured images of samples containing two-fluid phase and three-fluid phase systems were analyzed to obtain the water and LNAPL saturation (S_w and S_o). The R^2 results vary from 0.766 to 0.986 for the average OD and fluid saturation linear relationship. The distribution assessment of the LNAPL (isoparaffin liquid) showed that it was easily mobilized downward by the rainfall recharge. The recharge significantly reduced the LNAPL saturation at the upper part of capillary interface. At the lower interface, lens of LNAPL was observed to form higher S_o and tends to flow horizontally towards the water wells. This evaluation showed that rainfall recharge has significant effect on the LNAPL distribution.

The LNAPL spill containing benzene, which has similar properties to the one tested in the qualitative experiment was simulated using the MOFAT program. Simulations were performed for three different spill sites in a 2-D model domain. The results showed that different locations of spill site produced different shapes and levels of oil saturation contours. The predicted maximum oil saturation for the edge spillage and center spillage were 0.38 and 0.43, respectively. The distribution of oil saturation during LNAPL redistribution was influenced by the direction of the slope of water

table. The spill that occurred at the upper stream tends to create LNAPL lens along the groundwater surface if the higher LNAPL content was capable to move deeper reaching the groundwater level. Generally, 1 m^3 of oil spill containing 10.5% of benzene resulted in water and gas concentration ranging up to 183 g m^{-3} and 43 g m^{-3}, respectively. The distribution of concentration of water- and gas-phase was largely influenced by the direction of groundwater flow towards the lower water gradient.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

TAKSIRAN PROFIL AGIHAN CECAIR RINGAN FASA BUKAN AKUES DALAM ZON TAK TEPU DI BAWAH PENGARUH IMBUHAN HUJAN

Oleh

SAMIRA ALBATI BINTI KAMARUDDIN

Julai 2012

Pengerusi: **Profesor Madya Wan Nor Azmin Sulaiman, PhD**

Fakulti: **Pengajian Alam Sekitar**

Kebocoran dari simpanan bawah tanah dan tumpahan permukaan sumber hidrokarbon boleh menyebabkan pencemaran serius cecair ringan fasa bukan akues (LNAPL) dalam persekitaran subpermukaan. Dalam keadaan sebenar, aliran berbilang fasa semasa pergerakan LNAPL boleh dipengaruhi oleh imbuhan hujan. Untuk mempertimbangkan ini, satu kajian telah dijalankan untuk menyiasat agihan pergerakan LNAPL dalam zon tak tepu melalui ujikaji kualitatif dan kuantitatif, serta simulasi berangka. Kedua-dua ujikaji kualitatif dan kuantitatif menggunakan kaedah pantulan cahaya (LRM) untuk teknik pengimejan ketepuan NAPL.

Dalam ujikaji kualitatif, analisis imej menggunakan hubungan tentukuran lazim untuk menentukan profil agihan LNAPL dalam model 2-D. Kedalaman penembusan benzene dan toluene selepas 24 j suntikan dimulakan ialah masing-masing 37.0 cm dan 33.4 cm. Pada masa yang sama, kepulan toluene mengambil kawasan yang lebih besar (30.7%) berbanding kepada kepulan benzene (25.2%). Benzene bergerak lebih dalam seperti yang dijangka disebabkan oleh faktor perencatan, *R* yang lebih rendah

dan kelarutan air yang lebih tinggi berbanding toluene. Benzene lebih meruap kerana tekanan wap adalah lebih tinggi daripada toluene. Perbezaan ini menunjukkan bahawa sifat kimia sumber LNAPL mempunyai pengaruh yang besar ke atas mekanisme pergerakannya melalui media berliang. Imbuhan hujan menunjukkan kesan yang minimum kepada agihan benzene dan toluene disebabkan mekanisme pemeruapan dalam media berliang. Dalam ujikaji kuantitatif, teknik pengimejan ”multispectral” telah digunakan untuk membangunkan analisis imej yang boleh dipercayai. Purata ketumpatan optik (OD) daripada imej sampel yang diambil, yang mengandungi sistem fasa dua bendalir dan fasa tiga bendalir dianalisis untuk mendapatkan ketepuan air dan LNAPL (S_w dan S_o). Keputusan R^2 berbeza antara 0.766 hingga 0.986 untuk hubungan linear purata OD dan ketepuan cecair. Taksiran agihan LNAPL (cecair isoparaffin) menunjukkan bahawa ia mudah digerakkan ke bawah oleh imbuhan hujan. Imbuhan itu mengurangkan ketepuan LNAPL pada bahagian atas antara muka kapilari dengan ketara. Pada antara muka yang lebih rendah, kanta LNAPL terbentuk dengan S_o yang lebih tinggi dan cenderung mengalir secara mengufuk ke arah telaga air. Penilaian ini menunjukkan bahawa imbuhan hujan mempunyai kesan ketara ke atas agihan LNAPL.

Tumpahan LNAPL yang mengandungi benzene, yang mempunyai ciri-ciri yang sama dengan yang diuji dalam ujikaji kualitatif telah disimulasi menggunakan program MOFAT. Simulasi telah dijalankan untuk tiga tapak tumpahan yang berbeza dalam satu domain model 2-D. Hasil kajian menunjukkan bahawa lokasi tapak tumpahan yang berlainan menghasilkan bentuk dan tahap kontur ketepuan minyak yang berbeza. Ketepuan maksimum minyak yang diramalkan bagi tumpahan di pinggir dan tumpahan di tengah adalah masing-masing 0.38 dan 0.43. Agihan

ketepuan minyak semasa pengagihan semula LNAPL tertakluk kepada arah cerun aras air. Tumpahan yang berlaku di hulu aliran cenderung untuk membentuk kanta LNAPL sepanjang permukaan air bawah tanah jika kandungan LNAPL yang lebih tinggi mampu berpindah lebih dalam hingga ke aras air bawah tanah. Secara umumnya, 1 m^3 tumpahan minyak yang mengandungi 10.5% benzene menyebabkan kepekatan fasa air dan gas masing-masing sehingga 183 g m^{-3} dan 43 g m^{-3}. Agihan kepekatan fasa air dan gas sebahagian besarnya dipengaruhi oleh air bawah tanah yang mengalir ke arah kecerunan air yang lebih rendah.

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I certify that a Thesis Examination Committee has met on 16 July 2012 to conduct the final examination of Samira Albati binti Kamaruddin on her thesis entitled "Assessment of the Distribution Profile of Light Non-aqueous Phase Liquid in Unsaturated Zone Under the Influence of Rainfall Recharge" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctor of Philosophy.

Members of the Thesis Examination Committee were as follows:

Shaharin bin Ibrahim, PhD
Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Chairman)

Puziah bt Abdul Latif, **PhD**
Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Internal Examiner)

Mohd Razman bin Salim, PhD
Professor
Faculty of Civil Engineering
Universiti Teknologi Malaysia
(External Examiner)

Takeshi Katsumi, PhD
Professor
Kyoto University
Japan
(External Examiner)

_____
SEOW HENG FONG, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

Wan Nor Azmin Sulaiman, PhD
Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Chairman)

Mohamad Pauzi Zakaria, PhD
Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Member)

Norhan Abd Rahman, PhD
Associate Professor
Faculty of Civil Engineering
Universiti Teknologi Malaysia
(Member)

_____

BUJANG BIN KIM HUAT, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

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