Experimental and Numerical Simulation of Light Non-aqueous Phase Liquids Imbibition in Unconsolidated Porous Media

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1. Introduction

A three-dimensional multiphase laboratory experiment was simulated with a view to evaluating the subsurface distribution of lighter-than-water non-aqueous phase liquids (LNAPLs). The experiment was set-up to reconstruct the migration of hydrocarbon-leaks, occurring in the earth´s surface, predominantly driven by capillarity. Likely real-life occurrences and typical subsurface sources may include but not limited to, buried pipelines and underground storage tanks, from where leakages discharge. The study is, not only, targeted at understanding hydrocarbon capillary imbibition but intended to guide a sustainable approach towards limiting the spread of contaminants and in remediating contaminated subsurface system. Managing hydrocarbon subsurface contamination, would require solving problems associated with NAPL fate, flowrate, and extent of contaminants spread. The experimental outcome, validated by numerical results, provided the requisite model for resolving the above problems. A major consideration, with respect to decontamination of hydrocarbon leaks, is that the amount of oil released is always smaller than its catchment area, in terms of spreading and contamination. Richard's equation is applied in COMSOL Multiphysics® using equation-based interface to validate different laboratory experiments in order to model NAPL (e.g. hydrocarbons like diesel and crude oil) subsurface imbibition in porous media. It validated our empirical estimates suggesting that the porous media was not 100% saturated with fluids but are substantially occupied by air or gas. Our numerical model contribute solution for tracking the rate and extent of fluids spread in subsurface formation. The experimental study and numerical model can be utilized in simulating large-scale models in environmental related studies.

2. Experimental set-up

In Onaa et al (2020) and Onaa & Amro (2020), details of the experimental protocol for the various conceptual scenarios are presented. In this study, the experiment was setup in a way that made it possible for the fluid sample to enter the LNAPL reservoir, through an inlet pot fitted to the base-lid of the column, from an external oil tank. That way, a constant fluid supply to the soil profile was maintained under capillary imbibition. The propagation of the fluids – crude-oil and diesel - along the capillary height at time variations, was continuously observed, measured, and recorded until steady-state was attained. The experiment conducted for this study considered, firstly, LNAPL migration in upwards direction from the location of the leak-source and was monitored over 56 days period. The laboratory model seeks to reconstruct real-life leakage events emanating from buried and/or damaged subsurface infrastructures. In a second experiment, a separate single column - 100 cm x 15 cm in height and diameter – had its center fitted with a 5mm diameter inlet port. The experimental model is an extension of the procedure, described above wherein the similar approach was followed in depositing the model-sand. A coiled tube with multiple perforations was positioned at the center of the column with one end connected to the column inlet port. This design allowed LNAPL supply from the oil tank through the inlet port and tubing to ensure that the fluid saturations from the tube perforations circulated evenly within the soil profile. To determine the penetration depth as a function of capillary height, *hc*, reached, a timer was started the moment the fluid established contact with the model-sand, at time $t = 0$ and the corresponding fluid drawdown in the oil-tank to assess the volume, *v*, imbibed, for time-lapse, $t_0 = t_n$. *n* is the duration of the experiment. The purpose is, firstly, to recreate a scenario where a portion of vertical soil profile has come under the invasion of LNAPL leak, from a horizontal direction. Secondly, the set-up allows us to better visualize in which direction – upwards or downwards – that the fluids are more diffused over 20 hours period.

3. Mathematical model

The following assumptions are proposed, to simplify the mathematical model and are thus applied for both 1D and 2D-symmetric model:

- The porous medium is homogeneous else will be specified.
- The acting fluids are non-volatile in all experiments.
- Only subsurface migration is considered else will be specified.
- Entrance pressure effect is neglected.

3.1. One-dimensional (1D)-model

With Darcy´s law, the process of imbibition in porous media can be described in one dimension using the relation:

$$u = -\frac{k}{\mu}\nabla p \qquad (1)$$

Where, u, is the Darcy velocity in the medium, k, is the medium`s permeability, μ is the dynamic viscosity of the invading fluid, and p, the pressure within the pore space.

If we correlate the length of LNAPL migration with capillary pressure with respect to upwards- or downwards-acting gravity, Darcy law can be rewritten as in Xiao et al. (2018):

$$\frac{dz}{dt} = -\frac{k}{\mu}\left(\frac{p_c}{z} \mp \rho g\right) \qquad (2)$$

Whereas, z, is taken as the height of the fluid in the porous medium, the $\mp$ sign and p_c describe the direction of gravity acting in the domain, and capillary pressure, respectively.

$$p_c = p_{nw} - p_w = \rho h_c g \qquad (3)$$

Eq. (2) can be solved easily using Comsol ODE module or MATLAB while considering p_c as constant to test the applicability of Darcy's law for such phenomena. The 1D model was tested on different subsurface migration experiments through a model-sand profile. Fig. 1 illustrates the results for sand-model containing 10% carbonate with diesel as the saturating fluid. Capillary pressure value was estimated from Eq. (3) and had the value of 1710 Pa.

The model produced a strong correlation, with good fitting between the experimental and simulation curves. The negligible difference between both trend lines at the curved part can be attributed to the wall effect: an increased porosity to fluid-flow between sand profile and the column wall, compared to reduced diffusion with the model-sand.

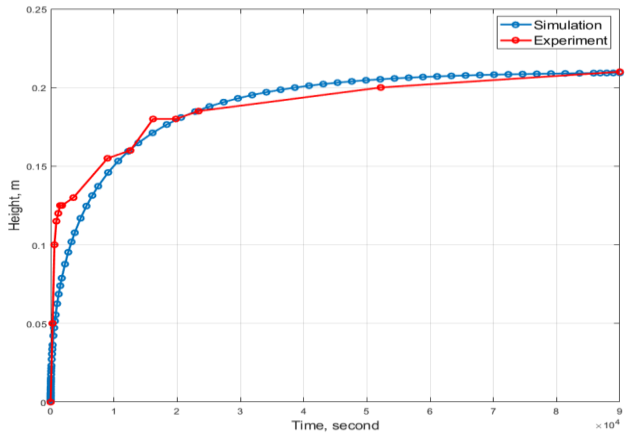

Figure 1. Schematic comparing experimental and simulation results of diesel subsurface imbibition in model-sand with 10% carbonate (Onaa et al, 2020).

3.2. Two-dimensional (2D) axisymmetric model

We propose a time and space-dependent saturation equation, with a view to clearly tracking the LNAPL saturation front. For the crude-oil and diesel fluids comprised in the porous system, a combination of continuity equation and Darcy law were solved simultaneously for both system-phases wherein either saturation or pressure, were considered as primary variable.

Considering wetting-(w) and non-wetting (nw) phase conditions, for a two-phase flow in porous system:

$$\varphi\rho_w\frac{\partial S_w}{\partial t} + \nabla\left[-\frac{kk_{rw}}{\mu_w}(\nabla p_w - \rho_w g\nabla z)\right] = 0 \qquad (4a)$$

$$\varphi\rho_{nw}\frac{\partial S_{nw}}{\partial t} + \nabla\left[-\frac{kk_{rnw}}{\mu n_w}(\nabla p_{nw} - \rho_{nw} g\nabla z)\right] = 0 \quad (4b)$$

With:

$$S_{nw} + S_w = 1 \qquad (5)$$

Following Hopmans et al. (1998), the above set of equations can be solved for saturation and pressure however, we take exception to relative permeability which, in our case, was not considered. A similar approach by Perez-Cruz et al. (2017), based on Richards' saturation equation, can be implemented as follows:

$$\frac{\partial S_w}{\partial t} = \nabla(D(S_w)\nabla S_w) + \frac{\partial K(S_w)}{\partial z} \qquad (6)$$

Here, S_w, is the saturation of the wetting-phase and $K(S_w)$ is hydraulic conductivity, while $D(S_w)$ which is the saturation-dependent diffusivity can be estimated from:

$$D(S_w) = \frac{k\lambda_w}{\varphi}\frac{dp_c}{dS_w} \quad (7)$$

Where λ_w considered as the motion of the wetting phase. Thus, we rewrite Eq. (6) in the following form:

$$\frac{\partial S_w}{\partial t} = \nabla(D(S_w)\nabla S_w) + \frac{\partial K(S_w)}{\partial S_w}\frac{\partial S_w}{\partial z} \quad (8)$$

Consequently, in the absence of experimental relative permeability data, $D(S_w)$ and $K(S_w)$ can be estimated (Perez-Cruz et al., 2017) for capillary pressure and relative permeability as a function of saturation accordingly:

$$D(S_w) = D_0\, {S_w}^m \quad (9)$$

$$K(S_w) = K_0\, {S_w}^n \quad (10)$$

With:

$$m = \frac{2\lambda + 1}{\lambda} \quad (11)$$

$$n = \frac{3\lambda + 2}{\lambda} \quad (12)$$

$$K_0 = \frac{k\rho_w g}{\mu_w} \quad (13)$$

We evaluated the terms, D_0 and λ, by simple trial and error method. It implies that the values were estimated by fitting our empirical data whereas adjustments between computational error and computational speed helped in selecting λ. Given that Eq. (8) is nonlinear and its nonlinearity being proportionate to the exponents, n and m, large values would require substantial simulation time. However, we would like to notify the appearance of certain undershooting signature in the curves - which required special treatment of the mesh and other technique called interpolation curve - in COMSOL Multiphysics. These may be observed in the results presented in next section.

3.3. Model implementation in COMSOL

To save simulation time, an axisymmetric domain is proposed, as illustrated in Fig.6. As can be seen, the mapped mesh is used instead of triangular mesh. Five elements are used in r-direction while 2000 elements were used in z-direction. The high-resolution mesh is required due to high nonlinearity of Eq. (8) and discontinuous initial and boundary conditions. These conditions triggered undershooting at all time steps, even at zero timestep, so a smoothing technique was used by applying step function in COMSOL. This step function was defined as the initial condition, in the domain, instead of zero value for saturation.

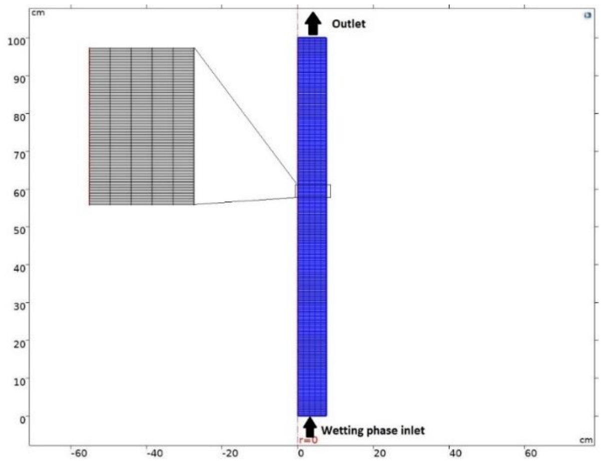

Figure 2. The axisymmetric domain with fine mesh.

Eq. (8) was implemented in COMSOL 5.5 in the form of partial differential equation (PDE) coefficient. Using COMSOL, Nassan and Amro, (2020) simulated immiscible two-phase flow in porous media and verified the applicability for fluid flow and heat transfer in porous media. Whereas the simulator is based on finite element, it efficiently defines models and post-processing results, through robust control capability offerings.

4. Results and Discussion

4.1. Results

Series of numerical experiments are implemented for different fluids (diesel and crude oil) to match all the lab experiments and the results illustrates a good matching.

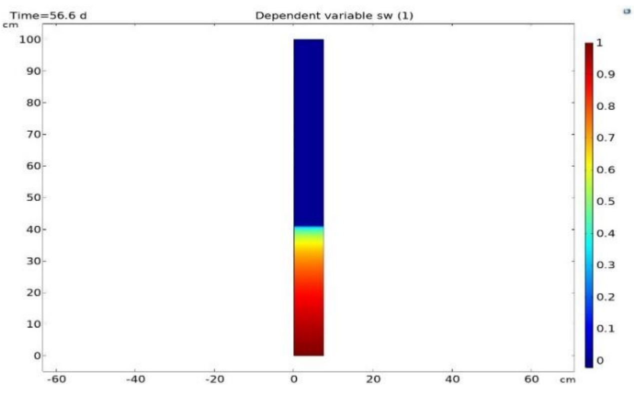

Figure 3. Two-dimensional model of the diesel saturation front in 56 days of imbibition.

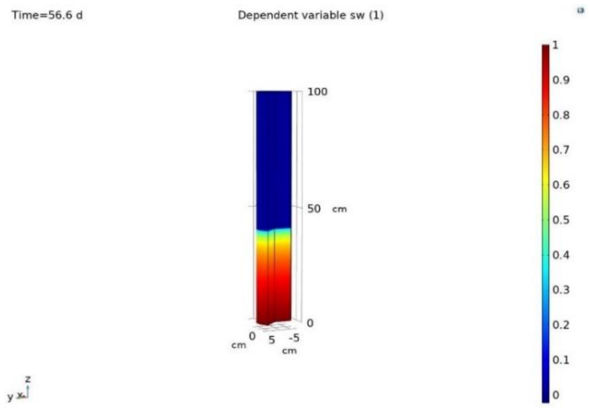

Figure 4a. Three-dimensional model of diesel saturation front in 56 days of imbibition.

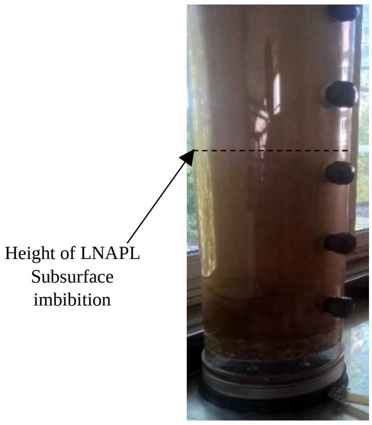

Figure 4b. Laboratory model illustrating diesel saturation front in 56 days of imbibition.

However, numerical results of LNAPL mobility presented in this study are predominantly diesel fluid, since it is established that the lighter components are responsible for the most contamination problems. Experiments in 2D axisymmetric, 3D, and the saturation propagation of diesel at different time steps, are presented in Fig. 3, Fig.4, Fig. 5. Saturation curves in 1D simulate the saturation at the centre of the column.

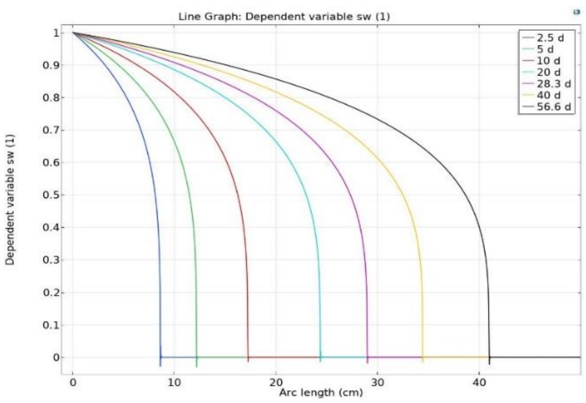

Figure 5. Plot of diesel saturation in 56 days of imbibition.

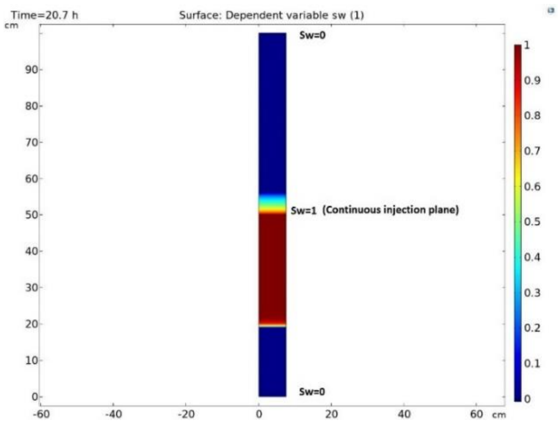

Figure 6a. Two-dimensional model of the diesel saturation front in 20-hours of imbibition. LNAPL injection was implemented at the middle of the column, to assess the direction with most oil migration.

Figure 6b. Laboratory model illustrating diesel saturation front in 20-hours of imbibition implemented as mid-column experiment.

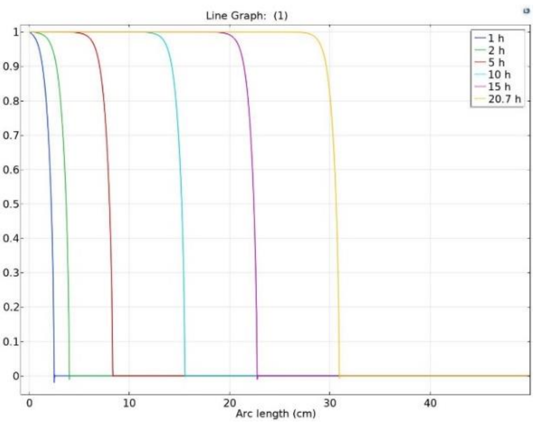

Figure 7. Plot of saturation model showing downward diffusion of diesel after 20 hours of imbibition.

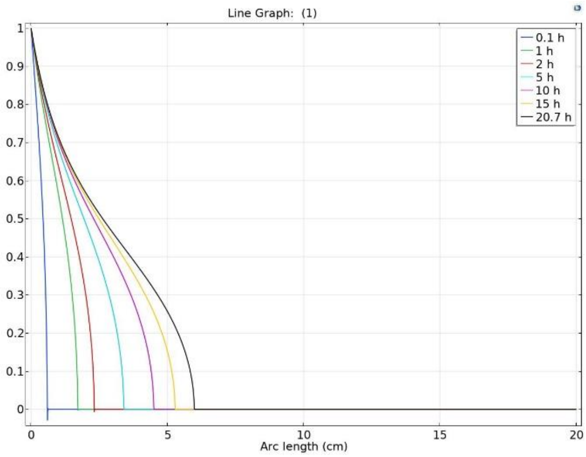

Figure 8. Plot of saturation model depicting upwards diffusions of diesel, after 20 hours of imbibition

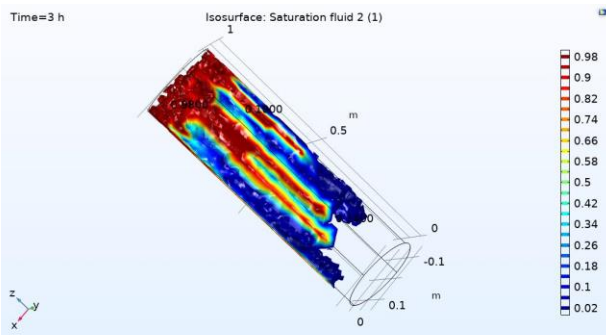

Figure 9a. Simulation model illustrating crude-oil Fingering and vaporization behaviour.

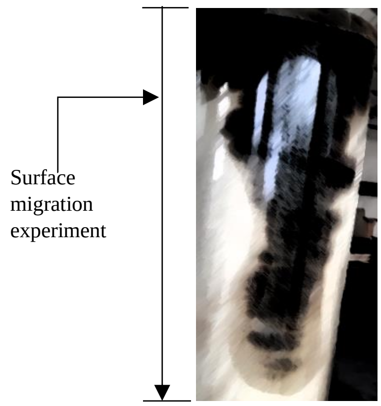

Figure 9b. Laboratory model illustrating crude-oil Fingering and vaporization behaviour.

4.2. Discussion

The numerical models presented in this study, has further demonstrated that COMSOL simulator retain the capability to estimate LNAPL subsurface imbibition in porous media. By implementing Richards equation (Eq.6) in COMSOL, the LNAPL diffusivity, D_0, and its rate, λ, were estimated, through trial and error, by fitting the experimental data. In order to choose, λ, there were trade-offs between computational error and computational speed. Given the nonlinearity of Eq. (8) which is seen to be proportional to the exponents, n and m, implementing higher values would require a significant simulation time. It is noteworthy to mention that multiphase system has a peculiar problem that was further revealed in the experiment. From the equations proposed by Pantazidou and Sitar, (1993), it was suggested that the NAPL lenses in the porous media may contain a considerable amount of water and air as opposed to the idea that these lenses are 100% NAPL. The study confirmed that the porous media was not 100% saturated with fluids but are substantially occupied by air or gas. The predictive charts presented from Figs. 3 to Fig. 5 are solutions for tracking the rate and extent of spread of diffusing fluids in subsurface formation. Saturation plots in Fig. 5 attests that the simulation model can predict crude-oil and other fluids distribution over-time and, at the same time, track the corresponding LNAPL subsurface location as can be seen in Figs. 3, and 4. The models yielded good correlation with the experimental results and are repeatable, for longer fluid migration forecasts. Again, the lighter hydrocarbon phase remains the culprit in the spreading of hydrocarbon contaminant.

When LNAPL leak occurs in the subsurface, bulk volume of the fluid would preferentially migrate downwards than in the upwards direction, due largely to gravity flow. This characteristic is depicted in Fig. 6, Fig.7, and Fig. 8. In LNAPL spill area, such as buried pipelines or storage facilities, fluids upwards-diffusion will stagnate slightly above the leak source while the sediment layers below the leak source will record greater penetration depth. But then, penetration depth in either direction, will depend on release amount and duration, since hydrocarbon partitioning and volatilization is ongoing within the vadose zone. Fig.9 illustrate LNAPL fingering and vaporisation behaviours. These flow behaviours facilitate LNAPL spread and environmental contamination. Thus, diffusion below the leak source, must be quickly contained to prevent contamination of groundwater system. The study outcome is thus, very strategic in tracking the fate, rate and location of LNAPL-based contaminants in the subsurface.

5. Conclusion

Accurate physics were described and mathematical model for the numerical simulation developed, based on the sediment and fluid data. Whereas the numerical model successfully tracked the LNAPL capillary imbibition and saturation fronts, its robust matching with the laboratory results was a good validation for our experimental model. Thus, the model sufficiently predicted the rate and location of leaking hydrocarbon fluids in the subsurface.

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