CENTRIFUGE MODELING OF LNAPL TRANSPORT IN PARTIALLY SATURATED SAND

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ABSTRACT: Model tests were performed at the Geotechnical Centrifuge Facility of Delft University of Technology, The Netherlands, to examine the mechanics of light nonaqueous phase liquid (LNAPL) movement in a partially saturated porous granular medium. The experiment simulated a 2D spill of LNAPL in an unsaturated sand prepared at two values of porosity. The duration of the centrifuge model tests corresponded to a prototype equivalent of 110 days. The choice of modeling a 2D flow together with the use of a transparent container enabled direct visual observation of the experiments. Scaling laws developed in connection with other centrifuge modeling studies were used to support the test results. Tests were conducted at two different centrifuge accelerations to verify, by means of the ''modeling of models technique,'' the similitude between the different experiments. The paper presents details of the experimental methodologies and the measuring techniques used to evaluate the final distribution of water and LNAPL content in the soils.

INTRODUCTION

The geoenvironmental damage caused by the release of liquids containing hazardous chemical substances into the subsurface is of major public concern. Predicting the transport of such contaminants in soil and ground water has proven to be an extremely challenging exercise. Mathematical and physical models have been developed to quantify such movements. A convenient and efficient physical modeling technique for these problems involves the use of a geotechnical centrifuge. One of the earliest applications of centrifuge modeling techniques to contaminant transport problems is due to Arulanandan et al. (1988). They showed that the length scale and modeling time can be reduced by subjecting the models to centripetal acceleration. This is due to the fact that the driving force to penetrate the contaminant increases with increased gravity while important dimensionless parameters are satisfied. Knight and Mitchell (1996) modeled the release of 1,000 L of (LNAPL) light nonaqueous phase liquid into an unsaturated fine sand. Their research described the influence of two different constant rates of release on the penetration of the LNAPL into the unsaturated granular medium. This paper describes the use of a centrifuge modeling technique to study the release of a LNAPL from a localized zone into unsaturated fine sand. The tests were carried out at the Geotechnical Centrifuge Facility (Fig. 1) located at Delft University of Technology, The Netherlands. The Delft University of Technology centrifuge has a diameter of 2.5 m, and samples with a weight of 300 N can be subjected to acceleration of up to 300 times the Earth's gravity field (Allersma 1994a,b). The experimental work described here extends the work of Knight and Mitchell (1996) by taking into account the influence of the porosity on the migration of the LNAPL into the unsaturated sand and its spreading within the saturated-unsaturated interface. The use of an LNAPL source that permits the time-dependent release

simulates a case of oil leakage from a storage tank. The choice of modeling a 2D flow coupled with the use of a transparent strongbox enabled direct visual observations of the oil infiltration. An LNAPL source that extends over the entire width of the container was used and the tests were conducted at two different accelerations. Scaling laws already developed by other authors (Arulanadan et al. 1988; Cooke and Mitchell 1991; Mitchell and Stratton 1994; Knight and Mitchell 1996) were used to establish the model dimensions. The influence of the capillary fringe and the water table on the LNAPL movement was investigated. Direct measurements of LNAPL content and water content are compared with images of the contaminant plume boundaries derived from the centrifuge experiments.

EXPERIMENTAL PROCEDURES

The experiments involved two aspects. First, an unsaturated profile was modeled by means of the centrifuge at both 20 and 30g. There, only one sand density was considered (dense sand, porosity equal to 35.9%). A bench test was also performed on a soil prepared at the same porosity. The objective of these tests was to observe the degree of similitude between the unsaturated profiles modeled using the centrifuge and that obtained under normal gravity. In the second part of the study, the partially saturated sand was used to model the LNAPL release from the line source. Two sand densities (porosity equal to 35.9 and 41.0%) in conjunction with two simulated gravities (20 and 30g) were used. The porous medium was Dutch dune sand with D_{50} equal to 0.20 mm, a coefficient of uniformity of 1.58, and placed at a dry density equal to 1,698 kg/m^3. The LNAPL used in the experiment was a motor oil having a density of 950 kg/m^3, a dynamic viscosity of 0.142 Pa·s, a vapor pressure of 0.01 kPa, and interfacial tension (air-oil) of 0.036 N/m. All of these properties were evaluated at a standard room temperature at 20°C. The ambient temperature during the operation of the centrifuge also corresponded to 20°C.

The container used in this research is shown in Fig. 2. The container consists of a metal frame on which two Perspex sheets are attached at a distance of 0.04 m apart. The internal dimensions of the container are given in Table 1. An outlet tap located at the base of the container permitted drainage of the sand sample in flight. The tap was opened manually just prior to the start of the centrifuge tests.

Two different LNAPL supply containers were used in the experiments to store the LNAPL. The dimensions of the containers were as follows: A 0.05 m × 0.03 m × 0.09 m container was used in the 20g test, and a 0.033 m × 0.03 m × 0.06 m container was used in the 30g test. The supply con-

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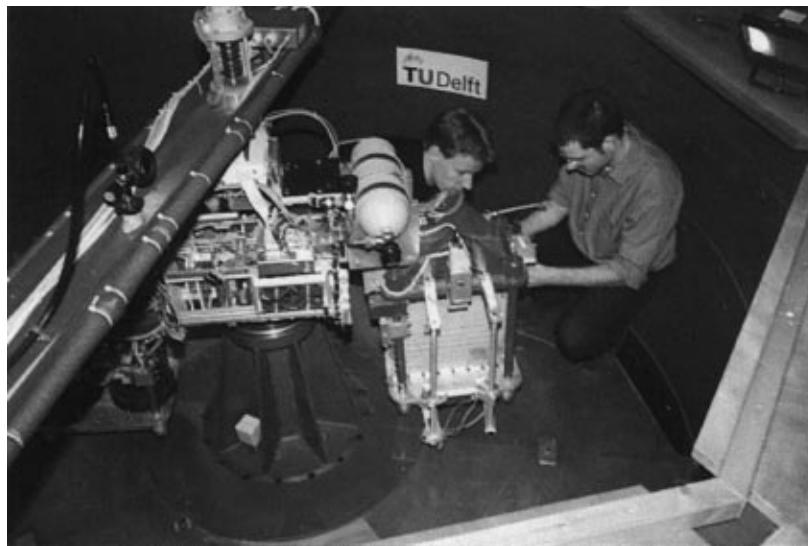
FIG. 1.   Centrifuge of Delft University of Technology

(a)

(b)
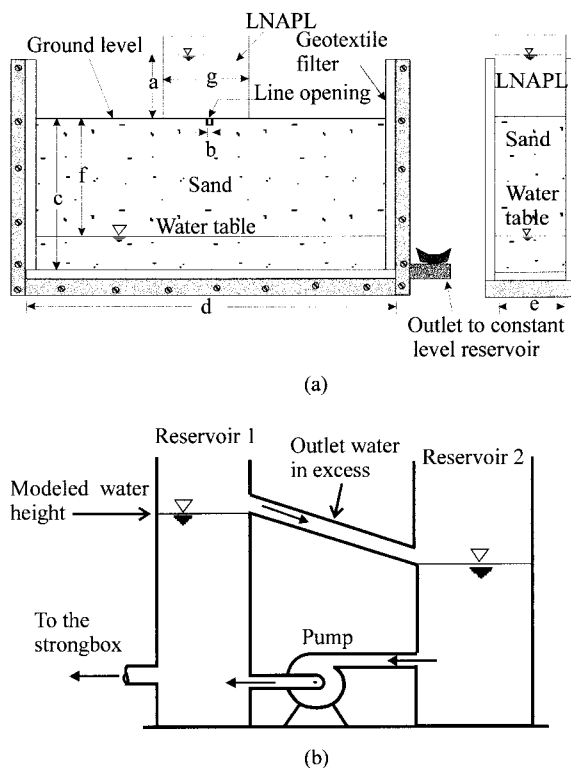
FIG. 2.   (a) Strongbox; (b) Constant Level Reservoir

FIG. 3.   Distribution of Moisture in Unsaturated Zone

TABLE 1.   Dimensions of Models

Model (1)	Dimensions (m)						
	a (2)	b (3)	c (4)	d (5)	e (6)	f (7)	g (8)
Centrifuge model 20*g*	0.083	0.0015	0.135	0.32	0.04	0.105	0.05
Prototype scale (20*g* model)	1.66	0.030	2.70	6.40	0.80	2.10	1.00
Centrifuge model 30*g*	0.056	0.0010	0.090	0.32	0.04	0.07	0.03
Prototype scale (30*g* model)	1.67	0.030	2.70	9.60	1.20	2.10	1.00

tainers were completely transparent so that it was possible to observe the decrease of the LNAPL level. Details about the centrifuge, the video equipment, and the sand model preparation are described by Allersma (1994a,b).

UNSATURATED PROFILE

Four models were used to study the distribution of water in the soil profile. Two tests were performed at 20*g* and two tests
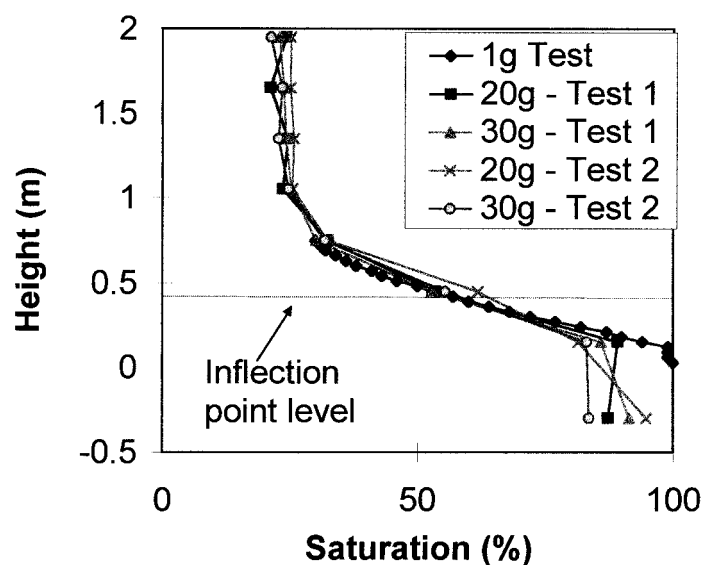
at 30*g*. Each test was performed twice to ensure the reproducibility of the results. Prior to conducting the centrifuge model tests, the capillary behavior was observed by performing a full-scale test. This 1*g* test was carried out by using two co-axial transparent Plexiglas cylinders. The internal cylinder had a diameter equal to 0.04 m and was cut into 25 equal rings 0.03 m in height. The assembled rings were contained in an external cylinder having diameter equal to 0.05 m. The base of the internal cylinder was closed by a porous fabric that prevented loss of sand. The experimental setup was prepared by raining the sand in the internal cylinder. Full saturation was obtained by performing the raining process under water. After compacting the sand to the desired density (35.9%), the setup was placed into a tank filled with 4 cm of water and the water in the cylinder was allowed to drain. After 4 days, the capillary fringe was stabilized and its height was about 0.42 m above the water level. After 15 days, the internal sample rings were then removed and water content was measured by drying at 125°C. Fig. 3 shows the measured distribution of saturation along the height of the sample. The inflexion point, which gives an indication of the boundary of the capillary fringe (Fetter 1993), is located approximately 0.4 m above the water table. This value is very close to the visual measurement made on the Plexiglas column. Thus, for water-wet soil, the use of

Plexiglas does not seem to influence the height of the capillary fringe in the soil.

The suction-moisture profile was prepared in the centrifuge by raining the sand in the centrifuge and by compacting it to the desired density. The model was then saturated by slowly injecting water through the valve of the container. Before commencing the tests, the outlet of the container was connected to a device that allowed keeping the head of the water constant throughout the experiment. The device, schematically shown in Fig. 2, consisted of two reservoirs placed on the swinging basket of the centrifuge. The first reservoir was connected to the tap of the container and had an outlet that kept the water level at the desired height. Such an outlet discharged the excess water into a second reservoir. The system was closed by a pipe that connected the second reservoir to the first reservoir passing through an electric pump working at 12 V. The electric pump provided a flow rate of 4.0×10^{-6} m³/s. The models, all prepared at a porosity equal to 35.9%, were drained at 20 and 30g for 54 and 24 min, respectively. On a prototype scale (N^2 scaling) this represents approximately 15 days. The progress of the drainage was monitored with an onboard camera. Optical measurement of the water content was carried out by using the image processing techniques described in Esposito and Allersma (1998). After 13 min at 20g and 6.5 min at 30g, the height of the capillary fringe was stabilized. At the end of the test, the sand models were extracted from the container and cut in slices 0.3 m thick (full-scale dimension), beginning from the surface of the sand. The mass of the water in the samples was determined by drying at 125°C. The abrupt stoppage of the centrifuge (less than 30 s) minimizes any redistribution of the water and ensures that the results are representative of values applicable to test conditions. In Fig. 3, the unsaturated profiles obtained for the four models are compared with that of the 1g test. In Fig. 3, the model dimensions are converted to prototype scale dimensions. The four profiles show a good agreement between each other, particularly in their correspondence within the unsaturated or vadose zone. The modeling of models procedure indicates that the unsaturated profiles obtained were reproducible. The four saturation curves for the centrifuge tests have a good agreement with the saturation curve for the 1g test. It should also be noted that the saturation curve of the 1g test is perhaps better defined due to the larger number of sampling points (every 3 cm) used to generate the curve. Despite the difference in detail, the inflexion point for all of the profiles is roughly at the same location (circa 0.40 m above the water table) of saturation, and the curves tend to the irreducible water saturation following the same trend. The major differences in the profiles are located in the saturated zone. This is attributed to the difficulty of determining the water content in the fully saturated zone with the procedure described above. After cutting the model in slices, part of the water content was lost. Preliminary measurements on saturated models showed that the loss of water could be as high as 5% in weight.

LNAPL RELEASE

The second part of the study consisted of simulating the leakage of a volume of LNAPL corresponding to a prototype equivalent of 1,660 L per meter of length. Two tests, at 20 and 30g, were performed on the sand strata prepared at a dense packing (35.9%) and at a loose packing (41%). The tests consisted of two parts: First, the sample was placed in the centrifuge completely saturated with water and allowed to drain for 54 min (20g) and 24 min (30g). The centrifuge was then stopped for no longer than 3 min and the LNAPL container was placed on the surface of the sand sample, pressed slightly into the soil, and filled with LNAPL up to the desired quantity. The centrifuge was again accelerated to the same value of

gravity and the LNAPL discharged into the sand. The discharge of LNAPL into the sand began before the centrifuge had reached the desired acceleration. Due to the low velocity of infiltration of the LNAPL into the sand (LNAPL saturated hydraulic conductivity equal to 1.72×10^{-8} m/s), and due to the short time required to bring the centrifuge to the desired acceleration (30 s to reach 30g), the start of the discharge was considered at the moment at which the centrifuge reached the required acceleration. The duration of the tests was 110 days (prototype time), which corresponded to 6.6 h at 20g and 2.9 h at 30g. The flow of LNAPL was monitored by means of the onboard camera. Because the container of the LNAPL was transparent, it was possible to observe the decrease in the level of the LNAPL with time (Fig. 4). Figs. 5–7 illustrate the progress of the LNAPL plume in the experiment with dense sand performed at 20g.

Comparing Figs. 6 and 7 it is evident that the ''leading edge'' behavior of the LNAPL plume is influenced by the presence of the capillary fringe. The LNAPL plume widened as it approached the region where the water saturation became close to 100%. In that region, the LNAPL was forced to migrate horizontally. The capillary fringe and the water table were displaced under the weight of the LNAPL phase. At the termination of the tests, water and LNAPL contents were determined. Each model was cut in slices and nine samples were taken every 0.3 m (prototype scale dimension) along the axis of symmetry of the plume. The tool used to collect samples was a sampler specially built for this research program. It consisted of a sampling tube attached to a guide rod. The sampling tube and the guide rod were contained in an external tube. A
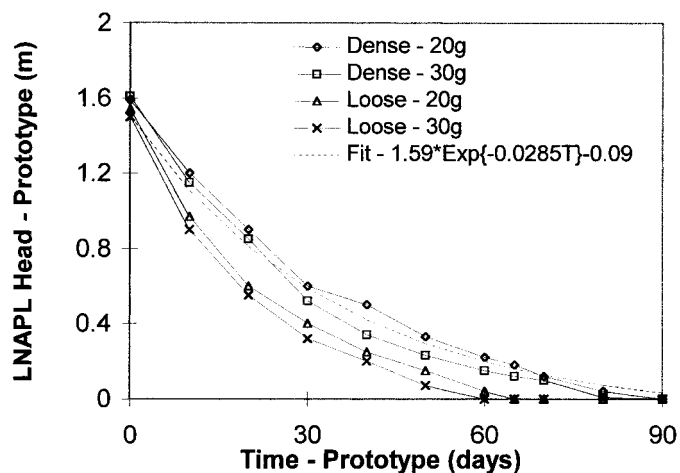

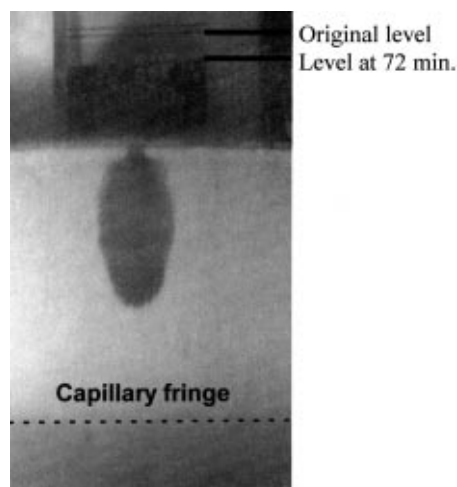**FIG. 4. Decrease in Time of LNAPL in Supply Tank**

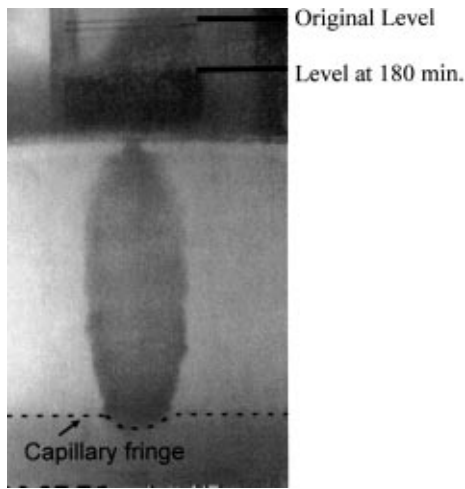**FIG. 5. Plume at Time = 72 Min (Prototype Time of 20 Days)**

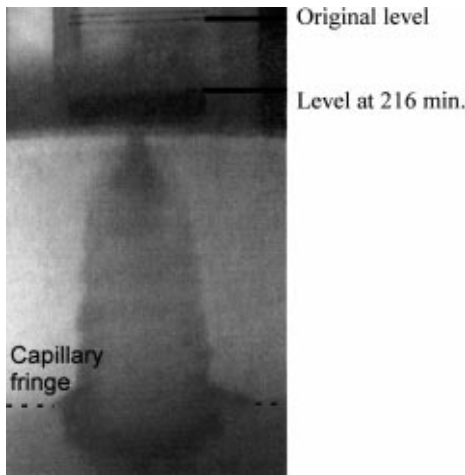**FIG. 6.   Plume at Time = 180 Min (Prototype Time of 50 Days)**

FIG. 7.   Plume at time = 216 Min (Prototype Time of 60 Days)

spring located at the top of the external tube made the sampling tube return to the initial position after sampling. A screw-nut located at the top of the external tube determined the length of the movement of the guide rod, and therefore, of the length of the sample extracted. The internal diameter of the sampling tube was 0.027 m for the sampler used for the 20*g* model, and 0.018 m for the sampler used for the 30*g* model. After oven drying at 125°C for 24 h, the LNAPL content was determined by washing the sand samples twice with gasoline. In this procedure, the sand was placed in glass containers, submerged into gasoline, and shaken mechanically for 30 min. The gasoline removed the LNAPL that coated the sand particles. The samples were then placed in a ventilated hood where the volatilization of the gasoline occurred. The weight of the samples was measured every hour for 24 h, with a balance having a sensitivity of ±0.001*g*, and the volatilization was considered complete when no variations in weight were recorded within a successive 8-h period. The difference between the initial and the final weight of the sand (after the second washing cycle) was attributed entirely to the weight of LNAPL in the sample. Previous tests showed that this technique was able to determine the LNAPL mass in the sand sample with an accuracy equal to ±0.005*g*. Fig. 8 shows the water and LNAPL content distribution at the prototype scale expressed as the ratio between the mass of the fluid and the mass of the solid.

There is good agreement between the tests carried out at different accelerations. Comparing the results for suction-moisture content profiles obtained in the first part of the experiment (Fig. 3) with the results shown in Fig. 8, it can be

seen that the presence of LNAPL decreased in the vertical extent of the capillary fringe and "depressed" the water table. In particular, the height above the water table of the inflection point of the moisture distribution curve moved from 0.42 m to less than 0.1 m. Fig. 7 shows that the water-LNAPL interface is located below the initial elevation of the water table. At the termination of the experiments, it was observed that some water flowed out of the strongbox. Thus, water was displaced by LNAPL and the water-LNAPL interface migrated below the initial water-table position because the column of free LNAPL exerted a positive fluid pressure on the water phase. For both models, the LNAPL content at the base of the sand was considered to be zero. It appeared, however, that the measured LNAPL content was small, but not zero (from 0.1 to 0.5% of the weight of the sand sample). This value could be attributed to the contamination of the sampler.

Each plume area was traced on the transparent sides of the container, scanned, and the data processed using graphics software. The final curves drawn on an identical scale are shown in Fig. 9 (dense sand) and Fig. 10 (loose sand). The presentation corresponds to prototype scale dimensions. The traces of the two plumes derived from the separate tests for the loose packing are quite similar. The two plumes derived from the separate tests on dense sand show a similarity with minor differences in the contours, particularly in the upper region.

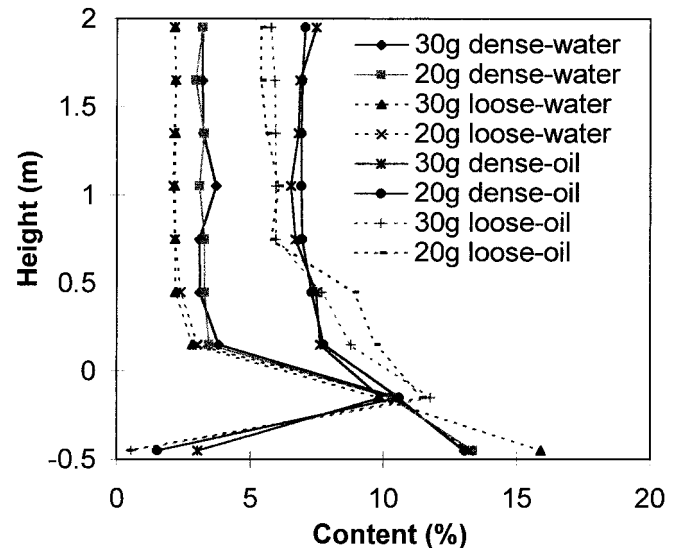For the loose sand, the time required to discharge all of the

FIG. 8.   Distribution of Water and Oil Content along Centerline of LNAPL Plume Measured at End of Tests (110 Days)
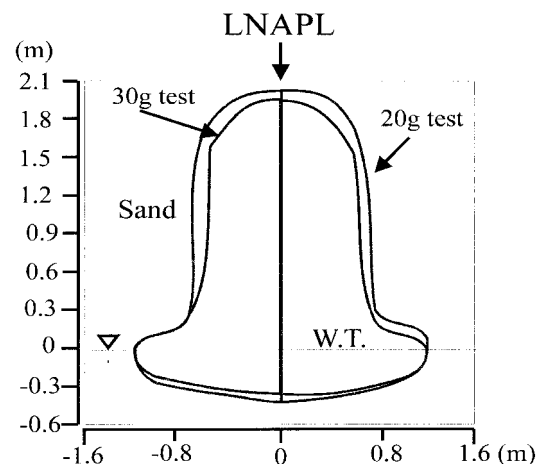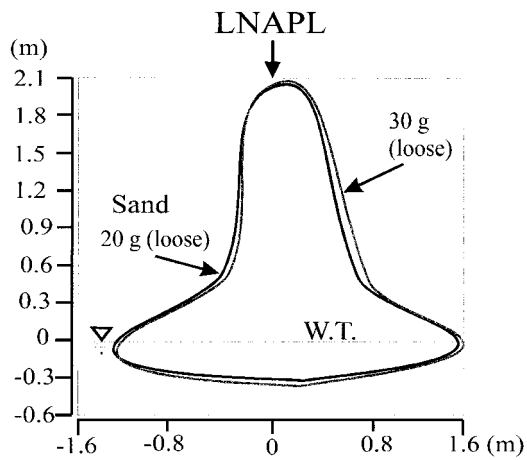

FIG. 9.   Boundary Lines of LNAPL Plumes at Termination (110 Days) of 20 and 30*g* Tests in Dense Sand

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FIG. 10. Boundary Lines of LNAPL Plumes at Termination (110 Days) of 20 and 30*g* Tests in Loose Sand

LNAPL out of the reservoir was shorter (Fig. 4), and the velocity of the advancing front of the LNAPL plumes was higher. Fig. 4 shows that, for each soil used for the modeling, the volume per unit time of LNAPL discharged into the sand is not affected by the *g*-level of the test, which confirms that the models are properly scaled. Knight and Mitchell (1996) modeled a discharge of LNAPL by using two different constant rates, namely, 319.68 L/day (high rate) and 143.42 L/day (low rate). The porosity of the sand they used can be estimated as 36%, and the saturated (with LNAPL) hydraulic conductivity close to 3.5×10^{-6} m/s (data not provided by the authors). In the study by Knight and Mitchell (1996), with a low release rate, when 1,000 L of LNAPL was discharged in 1 week, the front reached a depth of 3.2 m after 2 months. In this study, after 2 months, the total volume discharged was 1,128 L (Fig. 4) and the front reached the water table located 2.1 m under the LNAPL source (Fig. 6). The smaller infiltration recorded in this model was due to greater viscosity of the LNAPL (0.142 Pa·s for the LNAPL of this research and 0.0475 Pa·s for that used by Knight and Mitchell, 1996) and greater interface tension air-oil (0.0360 N/m in this research and 0.0215 N/m in the work of Knight and Mitchell 1996). Another difference between this study and the one of Knight and Mitchell (1996) is the methodology of discharge of the LNAPL. The plume obtained by Knight and Mitchell (1996) with low release has a maximum radius of about 1.5 m in the unsaturated zone. In the plumes modeled in this research, a dense sand (Fig. 9) showed a plume of a width in the unsaturated zone that is about the half of the value obtained by Knight and Mitchell (1996). Assuming that the hydraulic conductivity was isotropic in both sands used in the models, it seems that the rate of discharge is the factor that affects the extent of the movement of the LNAPL. The horizontal transport is favored with high release rates, whereas the vertical transport dominates at low release rates. When the injection rate of the LNAPL is at or above the saturated hydraulic conductivity, there is an excess of oil pressure promoting horizontal flow. At lower LNAPL release rates, there is a lower excess hydraulic pressure, and lateral flow is due mainly to suction gradients. Fig. 10 shows the traces of the plumes observed in loose sand. The width of the plumes in the unsaturated zone is about 0.8 m. Thus, with the same LNAPL column, an increase in porosity of 5% halves the width of the contaminated plume and reduces 23.5% of the time necessary to complete the spill (Fig. 4).

Fig. 8 shows that the modeled residual water content is about 2.5% in loose sand and about 3.5% in dense sand. The height of the capillary fringe, corresponding to the inflexion point on the water content profile (Fig. 8), is lowered from

0.42 m to less than 0.1 m for the dense sand model. In loose sand, buoyancy forces were favored to capillary and viscous forces, and the LNAPL could infiltrate vertically more easily as shown in Fig. 8. The factors that determine the microscopic mechanism of trapping can be expressed as dimensionless groups, known respectively as the Bond number N_B (ratio of gravity force to capillary force) and the capillary number N_{Ca} (ratio of viscous force to capillary force):

$$N_B = \frac{\Delta\rho g d^2}{\sigma}; \quad N_{Ca} = \frac{v\mu}{\sigma}$$

where $\Delta\rho$ = fluid density difference; g = acceleration due to gravity; d = particle radius; v = displacing fluid velocity; μ = displacing fluid viscosity; and σ = interfacial tension. At the same centrifuge acceleration, the Bond number was equal in both the dense sand model and the loose sand model, because the same materials were used. Thus, the ratio of gravity to capillary forces was similar in both models at the same acceleration level. To estimate the variation in capillary number, the velocity of infiltration of the LNAPL was measured by means of an image processing technique (Esposito and Allersma 1998). The velocity of infiltration of the LNAPL in loose sand was larger than that in dense sand. Consequently, the capillary number of the loose sand models was larger than that of the dense sand models. The capillary numbers calculated for the loose sand models were up to 1.19 times larger than those calculated for the dense sand models. Trapping in loose sand was, thus, smaller than trapping in dense sand (Morrow and Songkran 1981). The LNAPL accumulation on the water table was higher in loose sand than in dense sand. In contrast, the trapped amount of LNAPL in the unsaturated zone was higher in the dense sand than in the loose sand. The average LNAPL residual content ($\pm 0.1\%$) was 5.8% in loose sand and 6.9% in dense sand. The residual LNAPL content measured by Knight and Mitchell (1996) at a low LNAPL release at the center of the plume can be estimated to be about 5.9%. This result is extremely close to the residual LNAPL content measured for the dense sand model used in this study.

The estimation of the amount of LNAPL trapped in the porous medium can be influenced by the lack of similitude between centrifuge model and prototype (Knight and Mitchell 1996). The LNAPL concentrations of the 20 and 30*g* tests measured at the terminations indicate a similarity. Thus, by adopting the precautionary steps (low hydraulic conductivity of the porous medium, centrifuge acceleration up to 30*g*, a very small difference in density between wetting and nonwetting fluid), suggested by Knight and Mitchell (1996), it is possible to minimize the deficiencies associated with similitude of the LNAPL flow velocities resulting from different centrifuge accelerations.

OBSERVATIONS

After the completion of geotechnical centrifuge models, which simulated an LNAPL spill of 1660 L per unit length of the source extending over 110 days prototype time, the following can be summarized:

1. The LNAPL plume reached the water-saturated zone located at a depth of 1.6 m (prototype scale dimensions) below the ground surface displacing the capillary fringe and depressing the water table.
2. The spread of the plume was about 1.5 m in width (prototype scale dimensions) in the pendular zone for the tests involving dense sand, and about 0.8 m in width (prototype scale dimensions) for tests involving loose sand.
3. The measured residual LNAPL content in dense sand

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was about 5.8% and was comparable with the value measured by Knight and Mitchell (1996) using centrifuge tests on a similar porous medium and on an oil with a similar viscosity. An increase in porosity of 5% gave a residual LNAPL content equal to 6.9%.

4. Both with dense and loose packing, the LNAPL accumulated on the water-saturated zone possessed a sufficient positive pressure to displace the water table.
5. With the same porosity, the mode of discharge of the LNAPL influences the geometry of the contaminated volume, but it has no appreciable effect on the residual content of the LNAPL.
6. With the same mode of discharge, the porosity influences both the contaminated volume and the residual content of the LNAPL.

Furthermore, some general observations relating to the use of a centrifuge in modeling of multiphase transport phenomena can be made.

1. For loose sand models, the velocity of LNAPL discharge, measured fluid contents, and contaminated volumes appear to be similar at different g-levels.
2. The restrictions (i.e., low hydraulic conductivity of the porous medium, centrifuge acceleration up to $30g$, very small difference in density between wetting and nonwetting fluid), suggested by Knight and Mitchell (1996), are successful to achieve similarity. If the dimensionless parameters governing geotechnical centrifuge modeling of multiphase transport in an unsaturated zone are kept within a desirable range, similarity is not violated.

CONCLUSIONS

Centrifuge experiments were successfully used to observe the progress of an LNAPL plume resulting from release of the LNAPL at the surface of a porous granular medium. The prototype scale scenario corresponded to a deposit of Dutch dune sand resting on an impervious boundary located 2.7 m below the ground surface. The static ground-water surface was located 0.60 m above the base of the sand layer. In the prototype scale, the volume of the spill was 1660 L per unit length of the source and the duration of the simulations corresponded to 110 days. The simulation was carried out at two different values of gravity acceleration, so that the modeling of models procedure could be effectively employed. Furthermore, two different porosities of the sand were used in the centrifuge tests. This enabled the evaluation of the influence of porosity on LNAPL flow. The results of limited modeling indicate that porosity influences both the residual LNAPL content and the contaminated volume. Comparisons with the paper by Knight and Mitchell (1996) showed that the mode of discharge influenced only the extent of the contaminated volume. Results presented in this paper show a good correlation for different g-levels and confirm that centrifuge modeling applied to NAPL transport in soils can produce data and visual information for the purpose of validating computational models. Further confidence in the use of the results derived from centrifuge model tests can be gained by conducting large-scale tests in the laboratory or under field conditions.

ACKNOWLEDGMENTS

APPENDIX.   REFERENCES

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