

Iwona DESKA^{1*} and Katarzyna ŁACISZ¹

THE POSSIBILITY OF THE LIGHT NON-AQUEOUS PHASE LIQUIDS MIGRATION IN THE LAYERED POROUS MEDIUM

MOŻLIWOŚCI ROZPRZESTRZENIANIA SIĘ LEKKICH CIECZY ORGANICZNYCH W OŚRODKU POROWATYM O BUDOWIE WARSTWOWEJ

Abstract: The light non-aqueous phase liquids (LNAPLs) percolating into the soil and groundwater present a threat to the subsurface environment. When LNAPL is present on the groundwater table the initial step of remediation should be its recovery. Proper design and monitoring of LNAPL recovery require an estimation of the actual LNAPL thickness or LNAPL specific volume. These parameters are mostly calculated on the base of the apparent LNAPL thickness (LNAPL thickness measured in the monitoring well). This apparent thickness can be even several times greater than the actual LNAPL thickness in the porous medium surrounding monitoring well. This difference depends on the properties of soil and the properties and amount of LNAPL on the water table.

The major aim of this study was to investigate if LNAPL present in the observation well can contaminate the high permeable soil lenses lying below the LNAPL layer on the groundwater table. Results show that the considerable amount of LNAPL penetrated the high permeable soil lens that was hydraulically connected to the well. Additionally, the free product percolation into the high permeable soil lens has influenced the value of apparent LNAPL thickness. Such a situation can complicate the estimation of the actual thickness or specific volume of LNAPL on the groundwater table.

Keywords: LNAPL, actual thickness, apparent thickness, confined LNAPL, layered porous medium

The light non-aqueous phase liquids (LNAPLs) access the subsurface as a result of spills from the underground storage tanks and damages of pipelines and cisterns transporting fuels [1, 2]. A little amount of petroleum product infiltrates into the soil and groundwater with the leachates percolating from unsealed landfills [3] and due to the coal deposits exploitation [4]. Generally, migration of the LNAPL spilled into the porous medium comprises three steps [1, 5]: (a) vertical infiltration through the vadose zone due to the gravity and capillarity forces, (b) horizontal spread of LNAPL plume on

¹ Institute of Environmental Engineering, Faculty of Infrastructure and Environment, Czestochowa University of Technology, ul. Brzeźnicka 60A, 42-200 Czestochowa, Poland, phone: +48 34 325 09 17.

* Corresponding author: ideska@is.pcz.czest.pl

the groundwater table due to the buoyancy forces and relative permeability; it produces concurrent displacing of the capillary fringe; at this step LNAPL forms lens within the capillary fringe and some soluble constituents dissolve into the groundwater, (c) stabilization of the lateral spread of LNAPL plume and migration of dissolved contaminants in the direction consistent with the groundwater flow.

LNAPL comprises a special risk for soils and groundwater [2, 6]. When it is released into the subsurface it infiltrates downward through the unsaturated zone and can accumulate and float on the groundwater table. Then LNAPL can migrate laterally in the direction of groundwater flow [1, 7]. A portion of petroleum product can be adsorbed on the soil grains or entrapped in the voids of porous medium by capillary forces, posing the residual or entrapped LNAPL coexisting in the pores with water and air [8]. Mechanisms prompted the trapping of LNAPL in the porous media are: snap-off and bypassing [9, 10]. Snap-off mechanism prevails in pores that have high values of aspect ratio (pore body is much larger than the pore throat). Bypassing mechanism takes place when wetting fluid flow (water flow) disconnects the nonwetting fluid. In this case LNAPL ganglia are trapped in clusters of large pores [9–11]. In the water-wet saturated zone, the trapped non-aqueous liquid remains as disconnected and immobilized “ganglia” or “blobs”, referred to as the residual LNAPL (residual oil) [10, 11].

Mobile as well as immobile (residual) LNAPL is the source of soil and groundwater contamination [12]. It can act as long-term source of highly toxic substances [13]. LNAPLs usually consist of different substances (mixtures) with very diverse physical, chemical and biological properties [13].

Especially hazardous for groundwater resources are very soluble constituents originating from LNAPLs [13]. Polycyclic aromatic hydrocarbons (PAHs) are poorly soluble in water, but in specified conditions they can be leached out from contaminated rock and pose a threat to aquifer [14].

A certain part of LNAPL compounds evaporates easily due to high vapor pressures [13]. Both solubility and volatility of these constituents leads to the change of LNAPL composition and properties over time [13, 15].

The hydrocarbon contamination may influence the selected geological-engineering properties of clay soils. Clays contaminated with LNAPLs lose their plasticity and behave like a noncohesive soils [16]. This situation can lead to loss of insulating properties of clays and to the increase of risk of aquifer contamination [16]. The presence of LNAPL in the subsurface produces not only changes in soil morphology but also the disturbance of the biological processes on account of the pollution with toxic compounds [17].

The degree of risk for the subsurface environment associated with LNAPL spills is influenced by: the type and properties of contaminants (hydrocarbons), the volume of mobile LNAPL and its age, the migration pathways of contaminants, and the possibility of reaching and contamination of the water supply resources and systems [18, 19].

If the aquifer is contaminated with hydrocarbon, it is crucial that the remediation should be started as early as possible [20]. When sufficient amount of LNAPL is present on the groundwater table the initial step of remediation should be free product recovery.

After this process a certain amount of residual or trapped LNAPL remains in the soil pores due to capillary forces [20].

This immobile contamination can be removed from porous medium during further steps of remediation guided with use of the physical, physicochemical and biological processes [21]. The proper design of remediation strategy, requires the determination of the vertical and horizontal extent of the LNAPL body, the amount of LNAPL that can be recovered (mobile LNAPL), and estimation of the free product migration direction [2, 8, 20].

The free product volume is usually assessed on the base of thickness of LNAPL lens overlying the groundwater [19, 22] or on grounds of the LNAPL specific volume [22–24]. This thickness (the actual LNAPL thickness or the formation LNAPL thickness) or LNAPL specific volume can be assessed on grounds of the free product thickness measured in the observation wells installed in contaminated aquifer (the apparent thickness) [20]. Unfortunately, the apparent LNAPL thickness measured in the well is always different from the thickness of free product in the porous medium surrounding the borehole (the actual thickness) [18, 19, 22]. This difference depends on the hydrogeological properties of the aquifer and the amount and properties of released LNAPL [18]. Furthermore, the groundwater table in an observation well is always depressed by the overlying hydrocarbon plume and the LNAPL overlying the water in the porous medium surrounding the well may or may not penetrate through the capillary fringe and it is dependent on the flow rate through the unsaturated zone [19].

The notion of “apparent thickness” is relevant to conceptual model describing the vertical LNAPL distribution in the porous medium developed in the 1980s (a “pancake” model). This model assumes that LNAPL can spread horizontally on the groundwater table or on the capillary fringe forming the continuous separate layer. The “pancake” model didn’t take into account the phenomenon of capillarity [25]. This model assumes that almost whole pore space within the LNAPL lens floating on the groundwater table is filled with hydrocarbon so the LNAPL saturation within this lens amounts to nearly 100 percent [25].

The next conceptual model describing LNAPL distribution in the subsurface, developed in the 1990s is “the multiphase model” or “equilibrium model” (Fig. 1). This model is based on idealized relationships between capillary pressure and immiscible fluids saturation [22, 24, 25]. “The multiphase model” takes into account that water and air coexist with LNAPL in the pore space of contaminated zone of porous medium and LNAPL saturation do not reach 100 percent. The maximal LNAPL saturation amounts to approximately 70 percent [22, 25] and is observed near the water table [23, 25]. By “the multiphase model” the best parameter that enables estimation of recoverable free product volume is the LNAPL specific volume [22]. It is the volume of LNAPL per unit of area of soil surface [22].

Estimation of the actual thickness and the LNAPL specific volume requires the evaluation of the apparent LNAPL thickness [25]. The apparent thickness can be even several times greater than the actual thickness. Additionally, the lower limit of LNAPL layer in the well lies significantly below the lower limit of LNAPL layer in the adjacent geological formation [18, 22, 26, 27]. Thereupon the observation wells enabling the

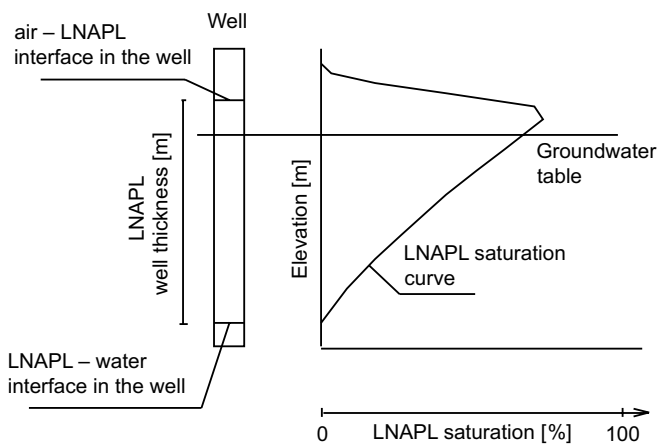


Fig. 1. LNAPL saturation curve in “multiphase model” (on the base of [25])

visibility of the whole LNAPL layer should be adequately deep. The presence of LNAPL in the observation well can be the result of presence of LNAPL on the groundwater table and its presence in the macropores (fissures, fractures) under the groundwater table which are in hydraulic contact with the LNAPL lens [28]. It is possible, that only a small part of LNAPL is present on the groundwater table, and the majority is located in the good permeable layer under the groundwater table having the hydraulic contact with LNAPL lens (so-called confined LNAPL) [28–30].

LNAPL layer can be confined within both: confined and unconfined aquifer [31]. The confined LNAPL in the confined aquifer can occur when LNAPL infiltrates into the classic confined aquifer, where high permeable soil layer is situated below an impermeable, semipermeable or low permeable porous medium (aquiclude, aquitard). In this situation LNAPL can migrate downwards through a low permeable zone within the network of higher permeable macropores, fissures, fractures, etc. The impermeable (semi- or low permeable) porous medium acts in this case as the confining layer [31].

On the other hand LNAPL can be confined in unconfined aquifer due to: lower historical water levels and LNAPL hydrostatic head and lateral migration. When the water table level in unconfined aquifer decreases LNAPL may infiltrate down to a coarser grained porous medium situated below the finer grained soil. The subsequent increase of groundwater table level may lead to submergence and confining of LNAPL within coarse grained porous medium [31]. Furthermore, the free product can penetrate some distance below the water table due to its hydrostatic head resulting from the LNAPL release. In this case LNAPL may access a coarser grained (high permeable) porous medium layer and migrate horizontally [31, 32].

The goal of experiment described in this paper is to examine how behaves the LNAPL layer in the observation well, that is located in layered, inhomogeneous porous medium and when the high permeable soil layer lies between low permeable soil layers. The additional goal of experiment was to confirm if such a situation may result in

secondary contamination of low permeable porous medium located above the high permeable soil layer.

Materials and methods

The experiment was performed with use of 3 soils including 2 types of sandy soils (soil 1 and soil 2) characterized in [33] and 1 type of gravel with grain diameter range from 5 to 20 mm. The effective grain sizes of soils 1 and 2 amounted respectively: 0.20 and 0.19 mm. Hazen's uniformity coefficients of soils 1 and 2 amounted respectively: 1.25 and 1.58 and their hydraulic conductivities amounted about 12.8 and 13.1 $\text{m} \cdot \text{d}^{-1}$.

As LNAPL was used the rapeseed oil with the density of 918 $\text{kg} \cdot \text{m}^{-3}$ and the dynamical viscosity of 0.07 $\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1}$ (at the temperature of 20°C).

The experiment was carried out in plexiglas column with diameter of 10 cm and height of 70 cm, with perforated tube having a semicircular cross-section as monitoring well (diameter of 3.5 cm). The filter wall was protected with nonwoven fabric to prevent the sand to get across the filter holes into the well. In the experimental column was also inbuilt the 3 cm diameter equalizing column with perforated bottom used to the changing of the water table elevations during the experiments.

The column was packed with the following layers of soils, starting at the ground surface: 0–32.4 cm – soil 1; 32.4–37.5 cm – soil 2; 37.5–43.5 cm – gravel; 43.5–54.0 cm – soil 1; 54.0–58.0 – soil 2; 58.0–64 cm – gravel. These values are the medium depths to the boundaries of specified layers, because these boundaries courses were not ideally horizontal. The good permeable layers (gravels) were separated from less permeable layers (sands) with use of non-woven fabric so that the sand does not pour into the gravel layer voids. The perforated tube for injection of LNAPL was located into the upper part of soil. Then the experimental column was filled with water until the water table reached the assumed elevation (below the outfall of perforated tube). After 4 days 100 cm^3 of LNAPL was injected into the column directly above the capillary fringe zone. LNAPL was stained with the dye – Sudan III for better visibility of the liquid in the column. After next 4–5 days the apparent LNAPL thickness was measured in the well. This procedure (LNAPL injection and consequent apparent thickness observation) was repeated 5 times (the volume of 5th portion of LNAPL was only 45 cm^3 on the grounds that the lower limit of LNAPL layer in the well declined near the bottom of column). The water table in equalizing column was kept constant during experiment and amounted 17.2 $\text{cm} \pm 0.1$ cm below soil surface. The top of experimental column was protected against LNAPL evaporation by the cover. Experiment was carried out at the temperature of 20 $^{\circ}\text{C} \pm 2^{\circ}\text{C}$ [33].

Results and discussion

Figure 2 shows the depths to upper and lower LNAPL limits in the well (the air-LNAPL interface and the LNAPL-water interface, respectively) for changing LNAPL volumes in the column.

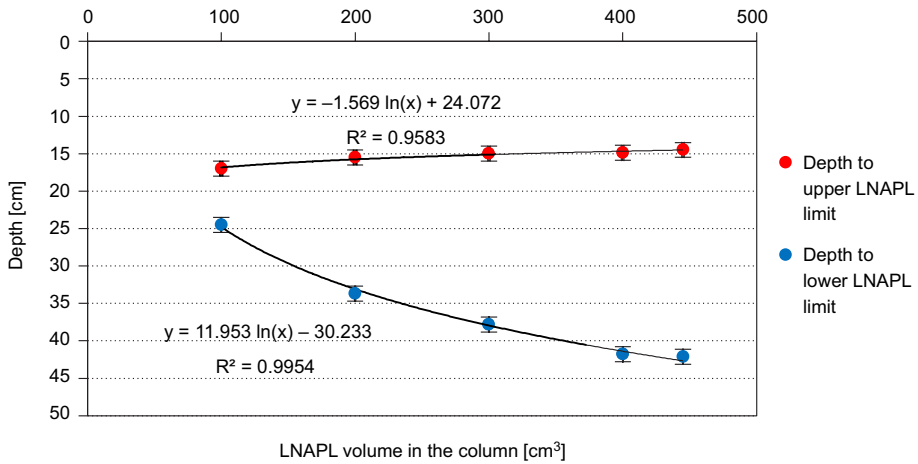


Fig. 2. Depths to upper LNAPL limits in the well (air-LNAPL interface) and lower LNAPL limits in the well (LNAPL-water interface) for changing LNAPL volumes in the column [33]

Figure 3 shows the changes of LNAPL apparent thicknesses depending on the LNAPL volume in the experimental column. Obtained results show that the biggest increases of the apparent LNAPL thickness were observed at the beginning of experiment, especially after the second LNAPL addition, when the thickness increase was $10.7 \text{ cm} \pm 0.4 \text{ cm}$ (for the apparent thickness $18.2 \text{ cm} \pm 0.2 \text{ cm}$). The thickness increases after third and fourth injections were distinctly lower and amounted to: $4.6 \text{ cm} \pm 0.4 \text{ cm}$ and $4.1 \text{ cm} \pm 0.4 \text{ cm}$, respectively. The least increase of apparent thickness amounted to $(0.7 \pm 0.4) \text{ cm}$ was observed after 5th addition of LNAPL (by 450 cm^3 of LNAPL in the column). Diminished increases of apparent thickness after 3rd and

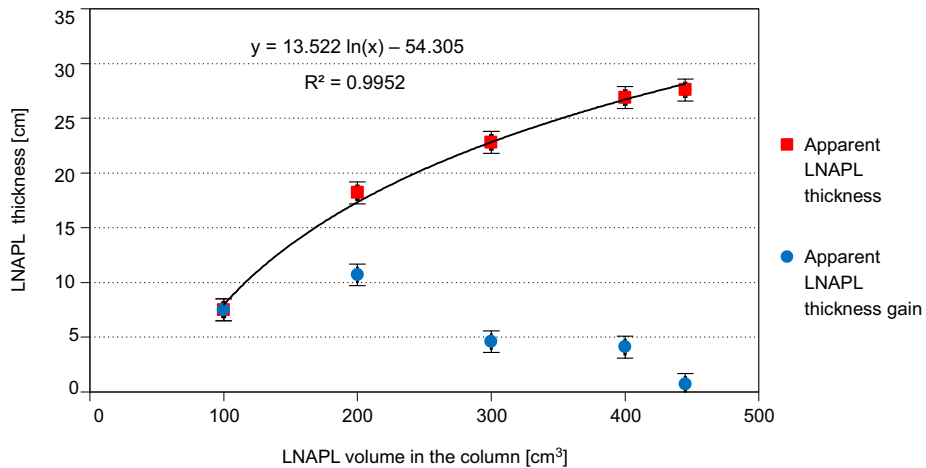


Fig. 3. Relationship between apparent LNAPL thicknesses and LNAPL volumes in the column (upper graph) and apparent thickness gains for changing volumes of LNAPL (lower graph) [33]

4th additions of LNAPL were caused by get of lower limit of LNAPL layer in the well to the upper high permeable layer of soil in column and filtration of LNAPL from well into the gravel layer (Fig. 4).



Fig. 4. LNAPL visible in the upper high permeable layer 1 day after its fourth addition (on the left) and 5 days after its fourth addition (in the middle and on the right [33])

Results presented in Figs. 2 and 3 show LNAPL layer distribution in the well a few days after each LNAPL addition. In the meantime, from the 3rd LNAPL addition, the lower limits of LNAPL layers observed just after LNAPL addition was located lower than the limits stabilized after a few days due to the slow LNAPL infiltration into the good permeable layer. Figure 4 shows that one day after 4th LNAPL addition oil is present in the well and in the whole good permeable layer (photo on the left). Five days after oil addition only the upper part of the gravel layer (about two-thirds of its pore space) was occupied by LNAPL and the lower part of gravel layer (about one-third of its pore space) was occupied by water. Initially, after 4th oil portion addition the apparent LNAPL thickness was a few centimeters greater than final value given in Fig. 3, what indicates the light residue on the column wall directly under the LNAPL layer (Fig. 4). Nonetheless, the excess of LNAPL little by little infiltrated through the high permeable layer into the low permeable layer, leading up with the apparent thickness decrease. On the other hand distinct retardation of the thickness increase after 5th LNAPL injection was caused not only due to the addition of the minor portion of LNAPL (only 45 cm³), but primarily due to the continuous LNAPL infiltration into the high and low permeable layers. As a result of this infiltration a few days after 5th LNAPL addition the lower limit of LNAPL layer raised and stabilised at the depth (41.8 ± 0.1) cm below soil surface (just as in the case of 4th LNAPL addition).

Further observations conducted about 2 months after the last LNAPL addition indicated that during this period occurred slow vertical upward LNAPL percolation from the upper high permeable layer (alternatively directly from the well) into the commanding weakly permeable layers composed of soils 1 and 2. In the end the

significant amount of LNAPL was immobilized near the water table and the capillary fringe level (Fig. 5).

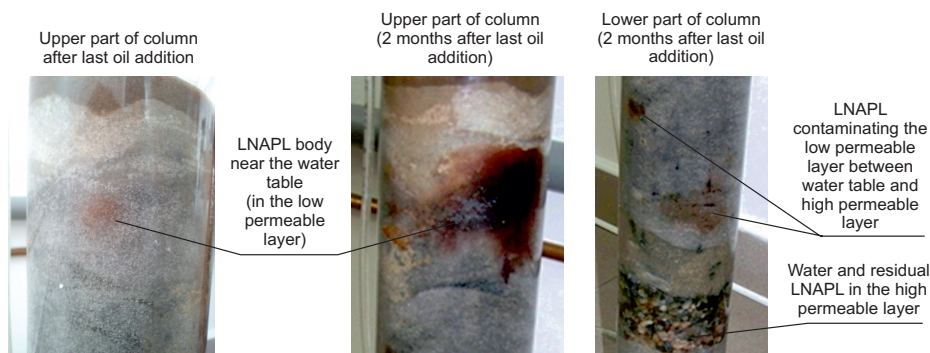


Fig. 5. LNAPL contaminating the low permeable layers 2 months after last addition of oil

The soil across which LNAPL percolated, located between upper high permeable layer and the water table, was distinctly contaminated with residual phase of oil. The source of this contamination was probably LNAPL present initially in the high permeable layer as confined LNAPL in the unconfined aquifer, described in [29–32]. However it is possible, that the source of soil contamination was also LNAPL infiltrating directly from the observation well. This means that there is a need for further more exact investigations to clarify LNAPL behavior in the subsurface, especially when LNAPL occurs in confined conditions. The aim of further research can be the investigation of the persistence of confined LNAPL in high permeable layer in the unconfined aquifer and its dependence on the grain size distribution of the overlying, water saturated soil layer.

Obtained results show that it exists real possibility of aquifer saturated zone contamination (even below the water table) when LNAPL present in the monitoring well reaches the high permeable soil layer. Additionally in this case the part of saturated zone located above the high permeable layer can be contaminated with LNAPL residual phase. The obtained results confirm that existence of high permeable layers having hydraulic contact with LNAPL layer present in the filtered well has an influence on the value of apparent LNAPL thickness and thereby on the accuracy of estimation of the actual thickness or specific LNAPL volume on the groundwater table and subsequent volume of LNAPL that can be recovered during initial remediation.

Conclusions

1. When the high permeable soil layer stays in the hydraulic contact with filtered well filled with LNAPL the light non-aqueous phase liquid can percolate into this layer.
2. The existence of high permeable layers having contact with the filtered well can have an influence on the apparent LNAPL thickness and thus on the accuracy of

estimation of the organic liquid volume that can be recovered during initial stage of remediation.

3. The results show that the considerable amount of LNAPL probably can percolate vertically from high permeable layers having contact with monitoring well into the commanding worse permeable layers of aquifer and can float towards the groundwater table.

4. Drilling of boreholes in the LNAPL contaminated aquifer can lead to the uncontrolled augmentation of range of the hydrocarbon contaminated zone. Therefore, after well drilling and installation the LNAPL recovery action should be began promptly.

5. LNAPL saturation depends on the soil grain size distribution. Coarse grained soil can accumulate more LNAPL than the fine grained soil.

6. There is a need for further investigations to clarify confined LNAPL behavior in the subsurface and to examine the persistence of confined LNAPL in high permeable soil layers in the unconfined aquifers.

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Instytut Inżynierii Środowiska, Wydział Infrastruktury i Środowiska
Politechnika Częstochowska

Abstrakt: Lekkie cieczy organiczne niemieszające się z wodą (LNAPL), które infiltrują do środowiska wodno-gruntowego, stanowią dla niego bardzo poważne zagrożenie. Gdy na zwierciadle wody podziemnej jest obecna warstwa LNAPL (tzw. wolny produkt), wstępny etap remediacji powinien obejmować jej szczypanie. Prawidłowe zaprojektowanie, a także monitorowanie operacji szczypania wolnego produktu wymaga znajomości rzeczywistej miąższości lub jednostkowej objętości LNAPL, które ustala się na podstawie miąższości LNAPL zmierzonej w studni obserwacyjnej (tzw. miąższości pozornej). Miąższość pozorna różni się od rzeczywistej i może być od niej nawet kilka razy większa. Różnica między wskazanymi miąższościami zależy m.in. od właściwości gruntu, a także od właściwości i ilości LNAPL na zwierciadle wody podziemnej.

Celem badań opisanych w artykule było ustalenie, czy wskutek obecności LNAPL w studni obserwacyjnej, zlokalizowanej w ośrodku porowatym o budowie warstwowej, może dojść do wtórnego zanieczyszczenia warstw dobrze przepuszczalnych, znajdujących się poniżej plamy LNAPL na zwierciadle wody.

Otrzymane wyniki wskazują, że część LNAPL może infiltrować ze studni obserwacyjnej do strefy ośrodka porowatego o wysokiej przepuszczalności, jeżeli pozostają one w hydraulicznym kontakcie. W takiej sytuacji może dojść do uformowania się tzw. napiętej soczewki LNAPL, która może występować zarówno w warstwie wodonośnej o zwierciadle napiętym, jak i w warstwie wodonośnej o zwierciadle swobodnym. Poza tym infiltracja LNAPL do warstwy dobrze przepuszczalnej może znacząco wpływać na zmniejszenie się miąższości pozornej, co może w dużym stopniu skomplikować prawidłowe ustalanie miąższości rzeczywistej i jednostkowej objętości LNAPL na zwierciadle wody podziemnej.

Słowa kluczowe: LNAPL, miąższość rzeczywista, miąższość pozorna, napięta LNAPL, ośrodek porowaty o budowie warstwowej