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Effects of alcohol-partitioning type and airflow on cosolvent flooding to benzene-LNAPL saturated porous media

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ABSTRACT

This study fundamentally investigated the swelling and distribution of benzene-light nonaqueous phase liquid (LNAPL) in porous media while cosolvent was flushed to the benzene-partially saturated system. Furthermore, the effects of simultaneous injection of cosolvent and air on the LNAPL behavior were visualized and thus quantified within a two-dimensional transparent porous medium. Partitioning types of alcohols affected dissolution of benzene entrapped in porous media. Tert-butanol (TBA) and 1-propanol floods apparently increased the LNAPL area, while a 70% ethanol flood reduced the LNAPL area by dissolution. Airflow facilitates mobilization of the swollen LNAPL by TBA and 1-propanol, while it facilitates dissolution of non-swollen LNAPL by ethanol. Therefore, LNAPL behavior during cosolvent flooding would be determined by partitioning type of alcohols and the presence of airflow.

1. Introduction

Spilled organic solvents and petroleum-based products to the subsurface are normally present as discrete liquid phases that are immiscible in water [1]. They are often referred to as nonaqueous phase liquids (NAPLs). Organic solvents that are heavier than water are referred to as dense nonaqueous phase liquids (DNAPLs), while liquids that are less dense than water are called light nonaqueous phase liquids (LNAPLs). NAPLs provide a continual source of dissolved constituents to the groundwater and become one of the most common groundwater contaminants [2,3].

Much effort has been made to remove NAPLs by using solubilizing and mobilizing agents [4–7]. Cosolvent (diluted alcohol) flooding has been suggested as an efficient technology for DNAPL source removal [7–11]. This method enhances DNAPL-dissolution by increasing the aqueous phase solubility of DNAPL or mobilizes DNAPL entrapped in porous media by decreasing the interfacial tension between the DNAPL and the aqueous phase. For an extension of cosolvent flooding, simultaneous injection of cosolvent with air, i.e., cosolvent-air flooding, was also suggested to improve a degree of contact between the cosolvent and the resident DNAPL [12,13].

Most cosolvent flood studies have used DNAPL as target organics to be removed from the porous medium. The cosolvent mixtures including alcohol are normally less dense than water and may exhibit override in porous media [14]. It is thus expected that this override may result in poor contact between cosolvent and the resident DNAPL. The poor contact between those aqueous phases leads to a low efficiency of cosolvent flooding for DNAPL removal. Due to the expected problem in DNAPL, cosolvent flooding may be appropriate for removal of LNAPL present in porous media. However, no attempt in the literature has been made to investigate cosolvent flooding for LNAPL removal. This study fundamentally investigated the swelling and distribution of LNAPL in porous media while cosolvent was flushed to the benzene-partially saturated system. Furthermore, the effects of simultaneous injection of cosolvent and air on the LANPL behavior were visualized and thus quantified within a two-dimensional transparent porous medium.

Three different alcohols, tert-butanol (TBA), 1-propanol and ethanol, were used for cosolvent flooding. These alcohols are known to exhibit different partitioning behavior in contacting with DNAPL [15]. This study expected that the partitioning behavior would affect the dissolution and mobilization of benzene within a porous medium. Therefore, the objectives of the study were to investigate the effects of partitioning type of alcohol on benzene removal from porous media, and to evaluate airflow impacts on cosolvent flooding.

2. Materials and methods

2.1. Experimental materials

This study used a glass porous media model to assess the spatial behavior of benzene entrapped in porous media. The transparent

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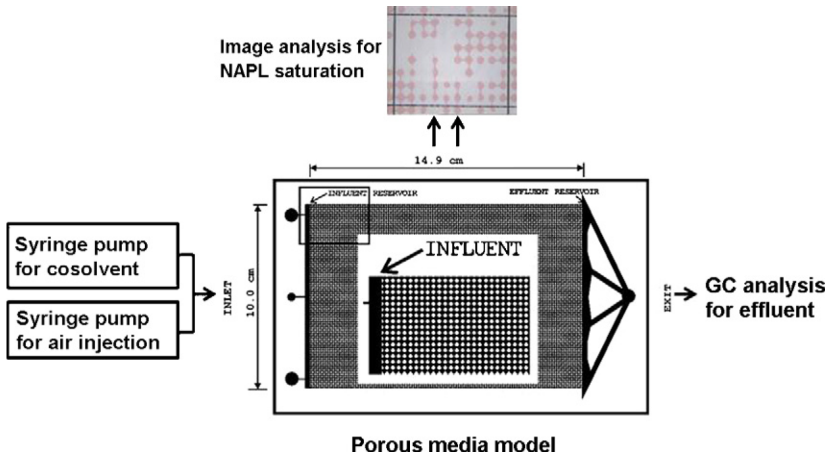

Fig. 1. Schematic representation of experimental setup.

two-dimensional porous media allow us direct visualization of benzene in a certain area. The transparent porous media was prepared by glass etching with a pore network pattern [12]. The pore network pattern had a length of 15 cm and a width of 10 cm. The pore volume and porosity of the porous media were 0.95 ml and 0.64, respectively. The permeability of the model was determined as $2.43 \pm 0.09 \times 10^{-8}$ cm^2. The average pore size of the network was 300 μm.

The experimental system consisted of two syringe pumps (Model 781100, KD Scientific Inc.) delivering fluids, a stereoscope (Zeiss SV11) visualizing microscopic behavior of benzene within the pore network, and an image analyzer (Optimas, Media Cybernetic Inc.) measuring the size of benzene blobs (Fig. 1). Similar types of experimental layout were already used for micromodel studies [12,13].

2.2. Experimental procedures

The porous media model was first flushed by carbon dioxide and then fully saturated with water. It normally took 1 day to fully saturate the model with water. Benzene (99%, Samchun Chemical, Korea) was chosen as target LNAPL for the study and was dyed with 0.5 g/l of Oil-Red-O (Sigma–Aldrich Co.) in order to distinguish it from the background. The benzene was injected into the porous media model through an injection port to reach the benzene saturation (the benzene volume to the void volume) at 0.10. The benzene-partially saturated model was then flushed by water at a flow rate of 2.6 ml/h to distribute the benzene and prepare it as disconnected NAPL blobs. Although the superficial velocity of water flooding corresponded to 8.6 cm/h, benzene blobs were still entrapped in the pores and those were placed over the upstream area of the model.

Three alcohols, TBA, 1-propanol and ethanol, were diluted into 30% TBA, 30% 1-propanol, and 70% ethanol by volume. Properties of the fluids used in the study are presented in Table 1. The aqueous phase density was determined by filling a known volume with the solution of interest and weighing it with an accurate balance (Model AR2140, Ohaus Corp.). The aqueous phase viscosity was determined using a viscometer (Brook field viscometer, Model LVDV-II+pro).

Cosolvent was injected into the benzene-partially saturated porous medium at a flow rate of 0.5 ml/h (a superficial velocity of 0.4 m/day). In the experimental condition of concurrent flow of

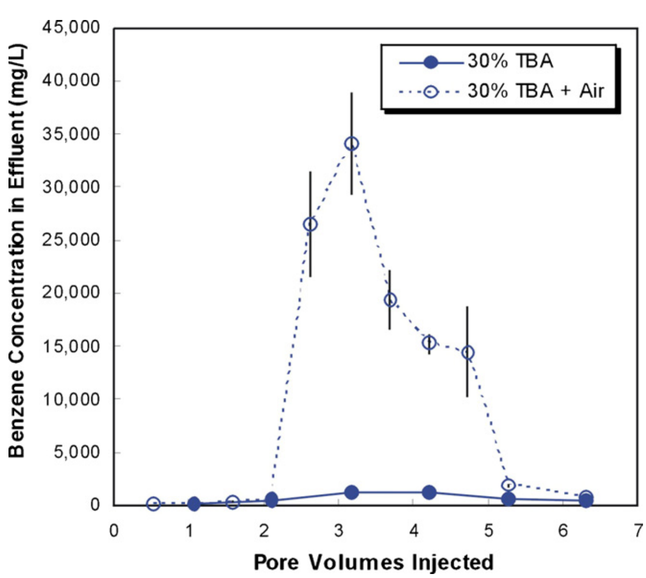

Fig. 2. Benzene concentrations of effluent discharged from the porous medium within which benzene NAPL was entrapped. 30% TBA was injected at a rate of 0.5 ml/h. In a case of '30% TBA + air', air was simultaneously injected with TBA at a rate of 1.5 ml/h.

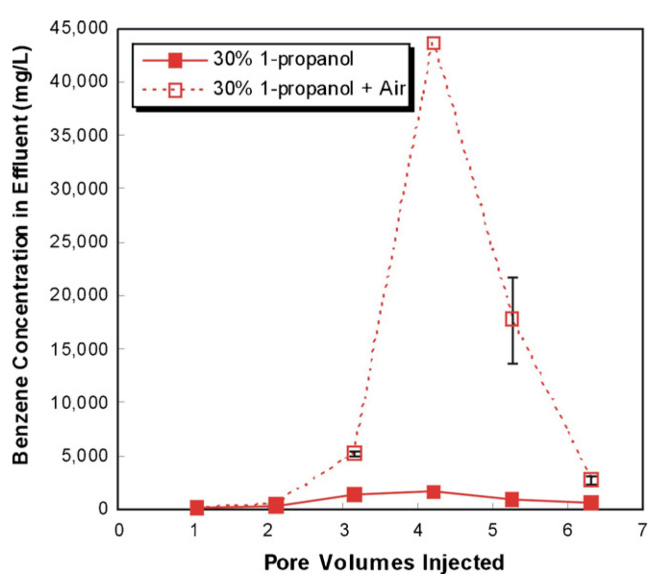

Fig. 3. Benzene concentrations of effluent discharged from the porous medium within which benzene NAPL was entrapped. 30% 1-propanol was injected at a rate of 0.5 ml/h. In a case of '30% 1-propanol + air', air was simultaneously injected with 1-propanol at a rate of 1.5 ml/h.

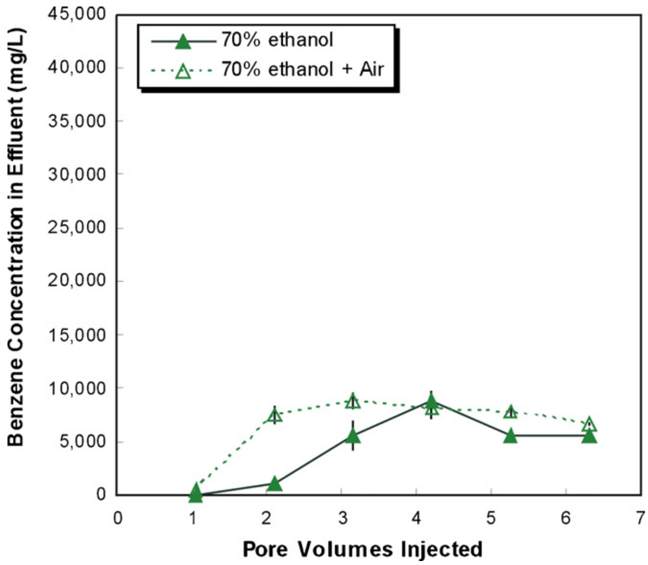

Fig. 4. Benzene concentrations of effluent discharged from the porous medium within which benzene NAPL was entrapped. 70% ethanol was injected at a rate of 0.5 ml/h. In a case of '30% 1-propanol + air', air was simultaneously injected with 1-propanol at a rate of 1.5 ml/h.

cosolvent and air, air was simultaneously injected from a syringe pump at a flow rate of 1.5 ml/h into the cosolvent flow.

Microscopic images were taken from the porous medium to investigate phase behavior. The 55 microscopic images taken from the two-dimensional porous medium were analyzed for the benzene area by using an image analyzer. The benzene concentration of effluent discharged from the porous medium was analyzed using an HP5890 Series II gas chromatograph equipped with a flame ionization detector and a Supelco SPB-1 capillary column. Benzene was extracted from the effluent samples by solid-phase microextraction (SPME, 65 μm polydimethyl siloxane-divinyl benzene fiber).

Table 1
Fluid properties[a].

Fluid	Density (g/ml)	Viscosity (cP)	Aqueous solubility (g/l)
Benzene	0.878[b]	0.65[b]	1.66
30% TBA	0.926	3.16	–
30% 1-propanol	0.928	2.26	–
70% ethanol	0.862	2.36	–

[a] Room temperature.
[b] 20 °C.

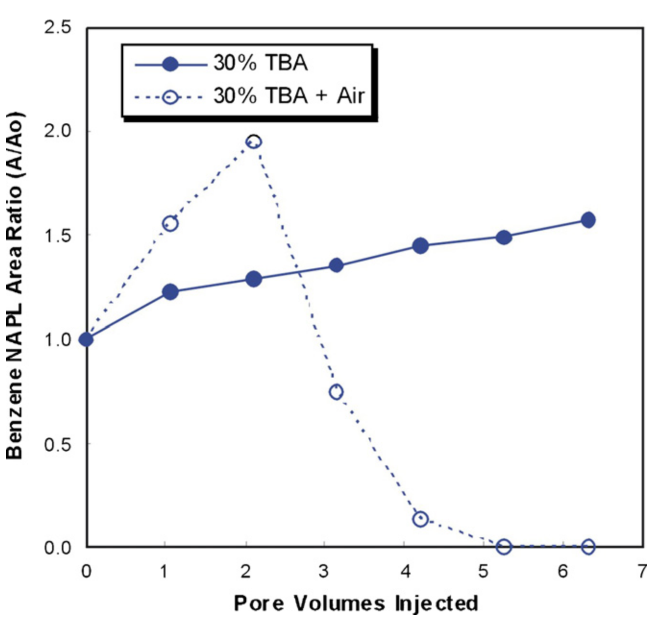

Fig. 5. Normalized benzene NAPL areas during 30% TBA flooding.

3. Results and discussion

3.1. Analysis of aqueous effluent under cosolvent flood only

Benzene concentrations in effluents collected from the benzene-partially saturated porous medium (10% of void volume was initially saturated by benzene) are shown in Figs. 2–4. The benzene concentration patterns of TBA flood (Fig. 2) are similar to those of

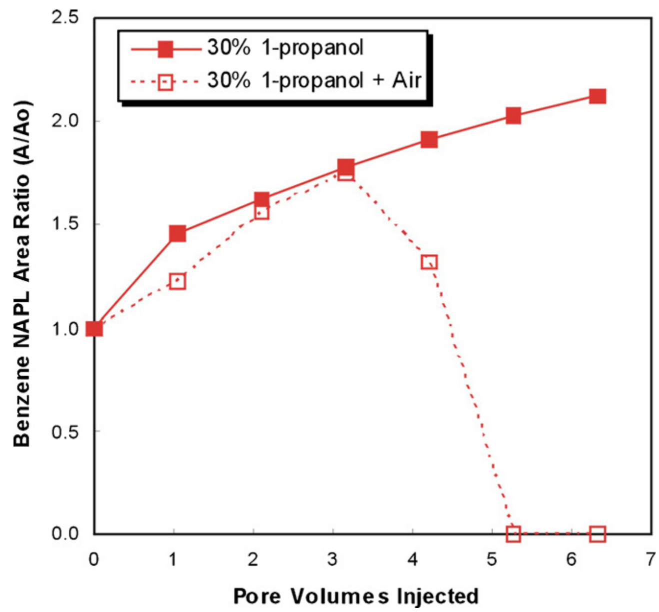

Fig. 6. Normalized benzene-NAPL areas during the 30% 1-propanol flood to a benzene saturated porous medium.

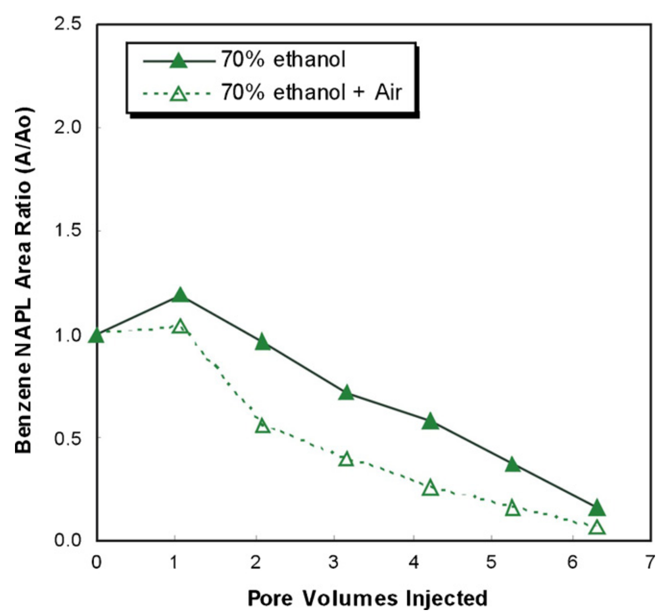

Fig. 7. Normalized benzene-NAPL areas during the 70% ethanol flood to a benzene saturated porous medium.

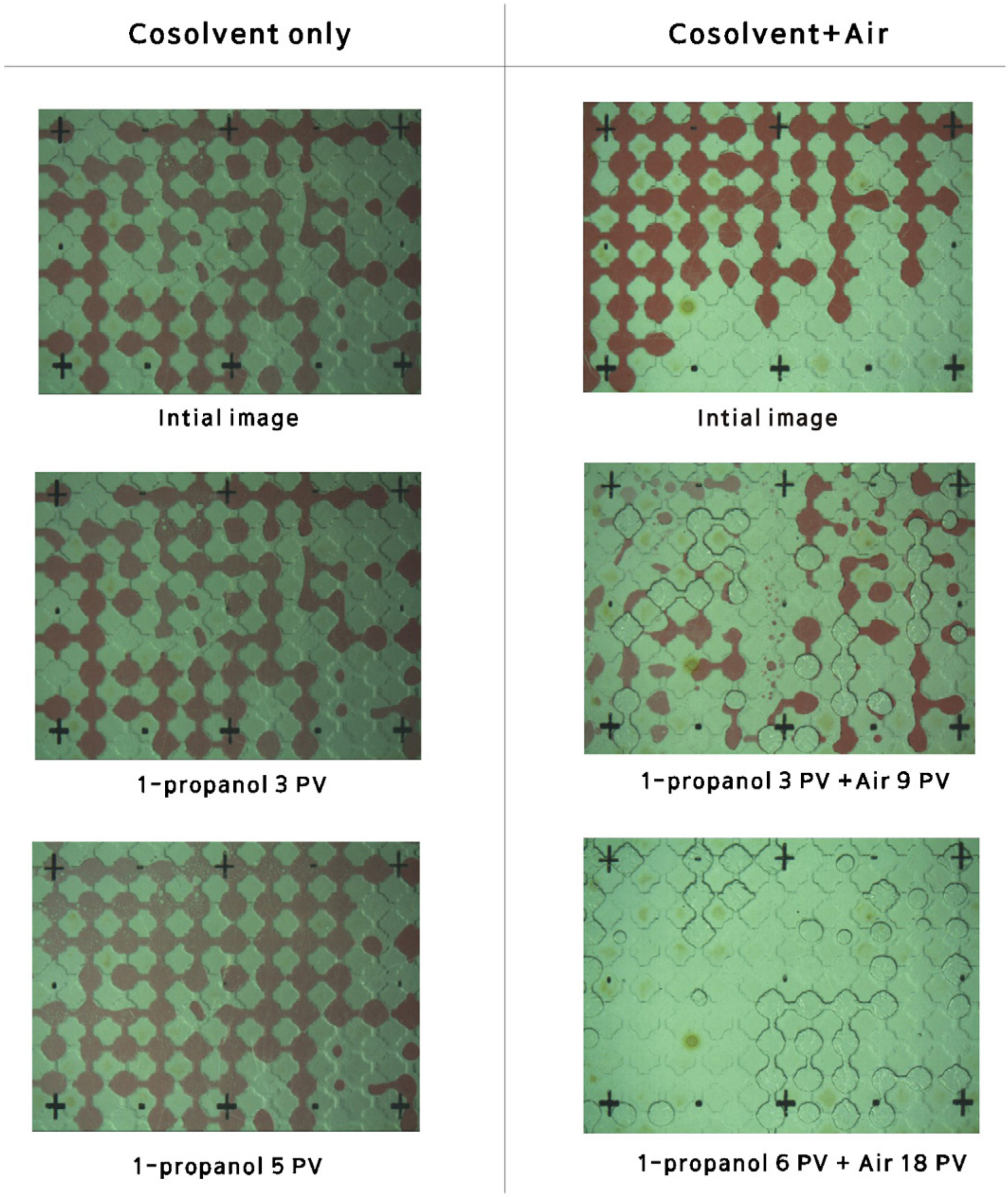

Fig. 8. Microscopic images taken from the two-dimensional porous medium; 30% 1-propanol was used for cosolvent.

1-propanol flood (Fig. 3). This section discusses cosolvent flooding only, while the next section discusses simultaneous flooding of cosolvent and air.

Benzene concentrations of 30% TBA and 30% 1-propanol floods were relatively lower than those of 70% ethanol flood only. Benzene concentrations of TBA and 1-propanol floods were around 1000 mg/l, while that of 70% ethanol flood showed greater than 5000 mg/l. The lower benzene concentrations in those flood effluents (30% TBA and 30% 1-propanol) would be attributed to the partitioning property of alcohol. TBA and 1-propanol preferentially partition into an organic phase, while ethanol preferentially partitions into the aqueous phase [16,17]. TBA and 1-propanol partitioning into DNAPL have been thus known as DNAPL-swelling alcohols, increasing the volume of the organic phase [18].

Alcohol partitioning behavior resulted in less dissolution of benzene. For those LNAPL-swelling alcohol floods, large amounts of remedial alcohol solutions would be required to completely dissolve LNAPL entrapped in porous media. Furthermore, there would be increased in the volume of LNAPL due to the cosolvent partitioning. The LNAPL volume increase is discussed in a later section.

Brandes and Farley [16] reported that TBA or isopropyl alcohol (IPA) content greater than 30% by volume was required for those partitioning into an organic phase. This study used 30% alcohol by volume. Preferential partitioning of alcohols into the benzene would occur under the experimental system. Therefore, the alcohol partitioning into LNAPL affects dissolution of LNAPL entrapped in porous media and determines a level of the effluent concentration.

3.2. Analysis of aqueous effluent under concurrent flow of cosolvent and air

Figs. 2 and 3 also showed that effluent benzene concentrations under concurrent flow of cosolvent and air (i.e., 30% TBA + air and 30% 1-propanol + air) were substantially higher than those of cosolvent flow only (30% TBA and 1-propanol). The results imply that airflow works to displace LNAPL blobs that were not mobilized when cosolvent flushed. Tiny LNAPL blobs were apparently

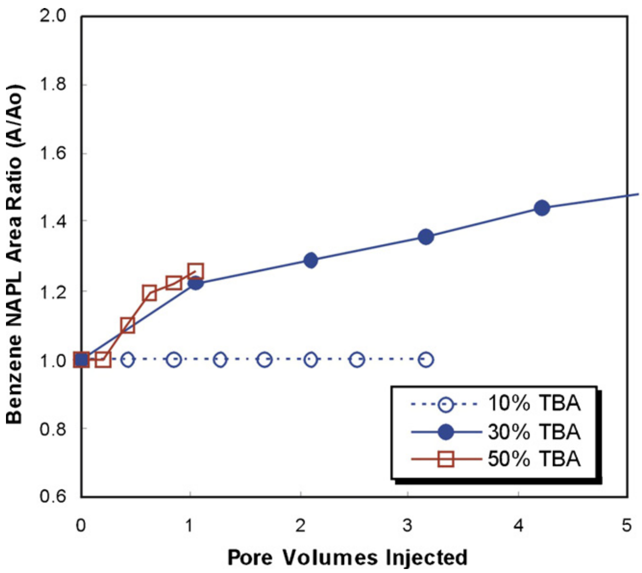

Fig. 9. Relative benzene NAPL area to the initial value under three different TBA concentrations.

discharged from the benzene-partially saturated porous medium, resulting in significantly high benzene concentrations. Airflow through the porous medium would displace the cosolvent and LNAPL blobs present at the pores. These immiscible displacement processes facilitated mobilization of LNAPL blobs through flow paths in porous media.

Contrary to the results of TBA + air and 1-propanol + air floods, an airflow impact on the ethanol flood was not observed as shown in Fig. 4. The maximum benzene concentrations were similar to each other (ethanol vs. ethanol + air). Most of LNAPL blobs would still reside in the pores under the ethanol and ethanol + air floods. Therefore, the major LNAPL removal mechanism by ethanol flooding would be dissolution of LNAPL rather than mobilization.

3.3. Quantification of benzene area in the 2-D porous medium

LNAPL-swelling of TBA and 1-propanol were clearly visualized and quantified directly from the transparent porous medium. Change in benzene area was monitored by using the image analysis technique already described in the experimental section. The total benzene area was determined from area measurements for benzene blobs. As shown in Figs. 5–7, the measured benzene areas (A) were normalized by the initial area (A_0) to produce the benzene area ratios (i.e., A/A_0).

3.3.1. Cosolvent flooding only

Benzene areas continuously increased as TBA and 1-propanol solutions were flushed. Those LNAPL swelling alcohols apparently increase the volume of benzene, which is explained by the increased benzene area measured from the transparent 2D system. However, 70%-ethanol flood showed a decrease in the benzene area after an initial slight increase as shown in Fig. 7. From those results, it is expected that TBA and 1-propanol may swell the LNAPL in the real contaminated site and result in an increase in source area contaminated by LNAPL.

In any flood employing alcohol solution only, mobilization of NAPL (i.e., both DNAPL and LNAPL) was rarely expected. Mobilization of NAPL occurs when the viscous force of remedial fluid is greater than the capillary force of the porous medium holing the NAPL. The ratio between those forces is expressed by the capillary number [19]. The flow rate of cosolvent solution was 0.5 ml/h. The capillary number at the condition was approximately 1×10^{-5}, which is much less than the critical capillary number of 10^{-3} for NAPL mobilization [20].

Fig. 8 (left column) shows microscopic images of cosolvent flooding. Swollen LNAPL was found in the image and remained for flooding time. TBA and 1-propanol floods increased the LNAPL area (see Figs. 5 and 6). TBA and 1-propnaol partitioned into the LNAPL and resulted in less LNAPL removal or less dissolution of LNAPL. Those results were consistent with the effluent concentration results mentioned earlier.

A 70% ethanol flood only reduced the benzene area (see Fig. 7). Therefore, the initial reduction in the benzene area can be attributed to dissolution of LNAPL into the aqueous phase.

3.3.2. Concurrent flow of cosolvent and air

All concurrent flow conditions resulted in decreases in the benzene areas just after the initial increase (see Figs. 5–7). The decrease in the benzene area indicates that the resident benzene was removed under the concurrent flow condition.

As shown in Fig. 8 (right column, the microscopic images of cosolvent + air flooding), air concurrently flowing with alcohol would displace the benzene that existed in the pores. Those immiscible displacement processes resulted in enhanced LNAPL removal from the porous medium. The major LNAPL removal mechanism under the concurrent flows of NAPL-swelling alcohols and air was the mobilization of LNAPL within the porous medium.

Contrary to the results of LNAPL-swelling alcohol, the NAPL area ratios involving 70% ethanol decreased as the cosolvent flushed (Fig. 7). The decrease in the NAPL area of 70% ethanol + air floods indicates enhanced dissolution of NAPL because significant mobilization of NAPL did not occur. As already shown in Fig. 4, relatively similar effluent concentrations were observed for ethanol and ethanol + air floods. This study showed that air flowing through the preferential flow paths of porous media displaces cosolvent from these paths into other flow paths, enhancing dissolution of LNAPL.

Quantification results of the NAPL area are consistent with the effluent concentration results already described in the previous section. Airflow facilitates mobilization of swollen LNAPL by TBA and 1-propanol, while it facilitates dissolution of LNAPL by ethanol. Impacts of airflow on NAPL behavior were clearly different according to the partitioning type of alcohol employed for a flood.

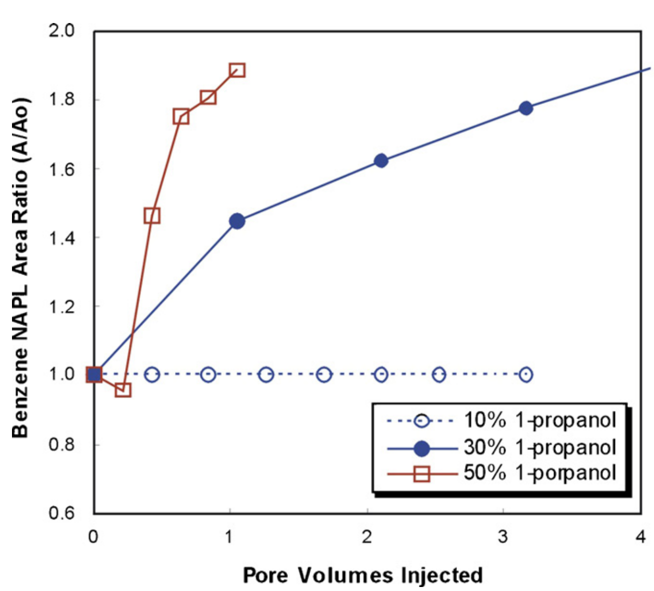

Fig. 10. Relative benzene NAPL area to the initial value under three different 1-propanol concentrations.

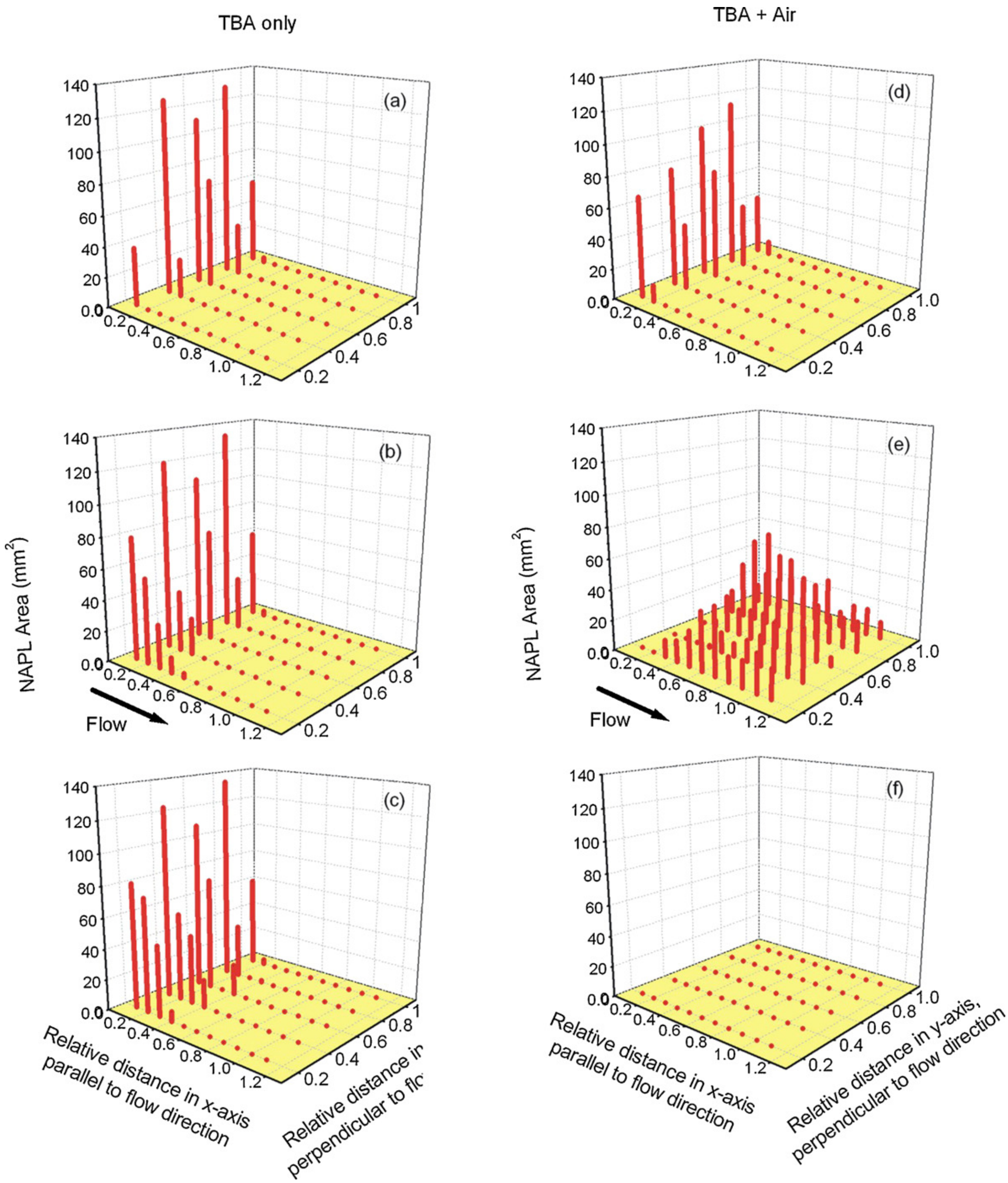

Fig. 11. Change in the benzene NAPL distribution on the two-dimensional porous medium under 30% TBA flow conditions: (a) initial NAPL distribution; (b) NAPL distribution after 4 h elapsed under 30% TBA flow; (c) NAPL distribution after 10 h elapsed under 30% TBA flow; (d) initial NAPL distribution; (e) NAPL distribution after 4 h elapsed under concurrent flow 30% TBA and air; (f) NAPL distribution after 10 h elapsed under concurrent flow 30% TBA and air.

3.4. Changes in the NAPL area by varying alcohol content

Figs. 9 and 10 show changes in the normalized benzene area by varying TBA and 1-propanol contents, respectively. At the relatively low alcohol content of 10%, the normalized NAPL area was constant for those alcohols. 1-Propanol more sharply increased the volume of benzene-NAPL while increasing the alcohol content. At least 30% alcohol content may be required for the partitioning of those alcohols into the NAPL. The results are consistent with the results of Brandes and Farley [16] that mention an alcohol content greater than 30 vol.% for partitioning into the NAPL. From the relative NAPL area results of Figs. 9 and 10, a degree of NAPL swelling would be well understood under the swelling alcohol flow conditions in porous media.

3.5. LNAPL distribution over the 2-D porous medium

3.5.1. Cosolvent flooding only

Figs. 11–13 display variations in the benzene distribution on the two-dimensional porous medium under 30% TBA, 30% 1-propanol, and 70% ethanol flow conditions, respectively. TBA and 1-propanol floods showed the similar benzene distribution patterns. Those LNAPL-swelling alcohols increased the area of LNAPL all over the porous medium (see Figs. 11 and 12(a)–(c)). However, the heights

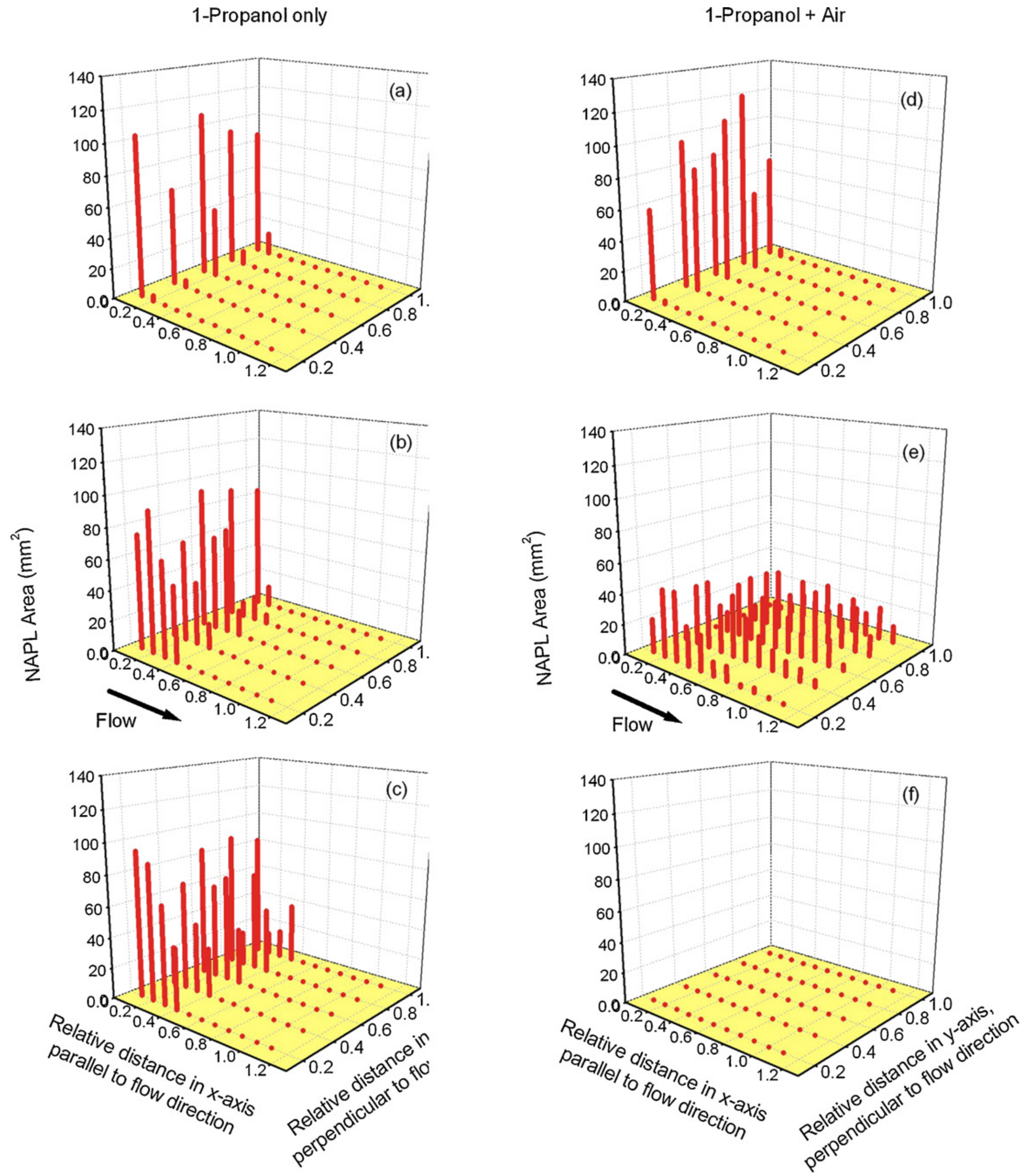

Fig. 12. Change in the benzene NAPL distribution on the two-dimensional porous medium under 30% 1-propanol flow conditions: (a) initial NAPL distribution; (b) NAPL distribution after 4 h elapsed under 30% 1-propanol flow; (c) NAPL distribution after 10 h elapsed under 30% 1-propanol flow condition; (d) initial NAPL distribution; (e) NAPL distribution after 4 h elapsed under concurrent flow 30% 1-propanol and air; (f) NAPL distribution after 10 h elapsed under concurrent flow 30% 1-propanol and air.

of the LNAPL area under ethanol flow conditions were decreased as ethanol was flushed (see Fig. 13(a)–(c)). Those results were completely consistent with the quantification results of the LNAPL area as discussed in the previous section.

A 70% ethanol flood only reduced the height of the benzene area over the porous medium, while TBA and 1-propanol floods increased the height of the LNAPL area. The reduction in the height of the benzene area can be attributed to dissolution of NAPL into the aqueous phase.

3.5.2. *The concurrent flow of alcohol solution and air*

Although TBA and 1-propanol floods showed that most of the swollen LNAPLs resided near the upstream region of the porous medium, LNAPL of the concurrent flow conditions were mobilized along the flow direction (see Figs. 11 and 12(d)–(f)).

Even in the ethanol + air concurrent flow conditions, the LNAPL were little mobilized toward downstream. However, the tendency of the mobilization was less than that of the swelling alcohols. Falta [15] also indicated that DNAPL under a swelling alcohol flow condition would be easily mobilized in porous media, compared to a non-swelling alcohol condition.

Those results were consistent with the previous effluent concentration and the total benzene area measurement results. The concurrent flow of alcohol solution and air resulted in mobilization of LNAPL, giving a rise in the benzene effluent concentration and significantly reduction in the benzene area. Airflow enhanced the mobilization of LNAPL for swelling alcohol floods (TBA and 1-propanol) and the dissolution of LNAPL for ethanol flood. Therefore, LNAPL behaviors during cosolvent flooding would be determined by partitioning types of alcohols and the presence of airflow.

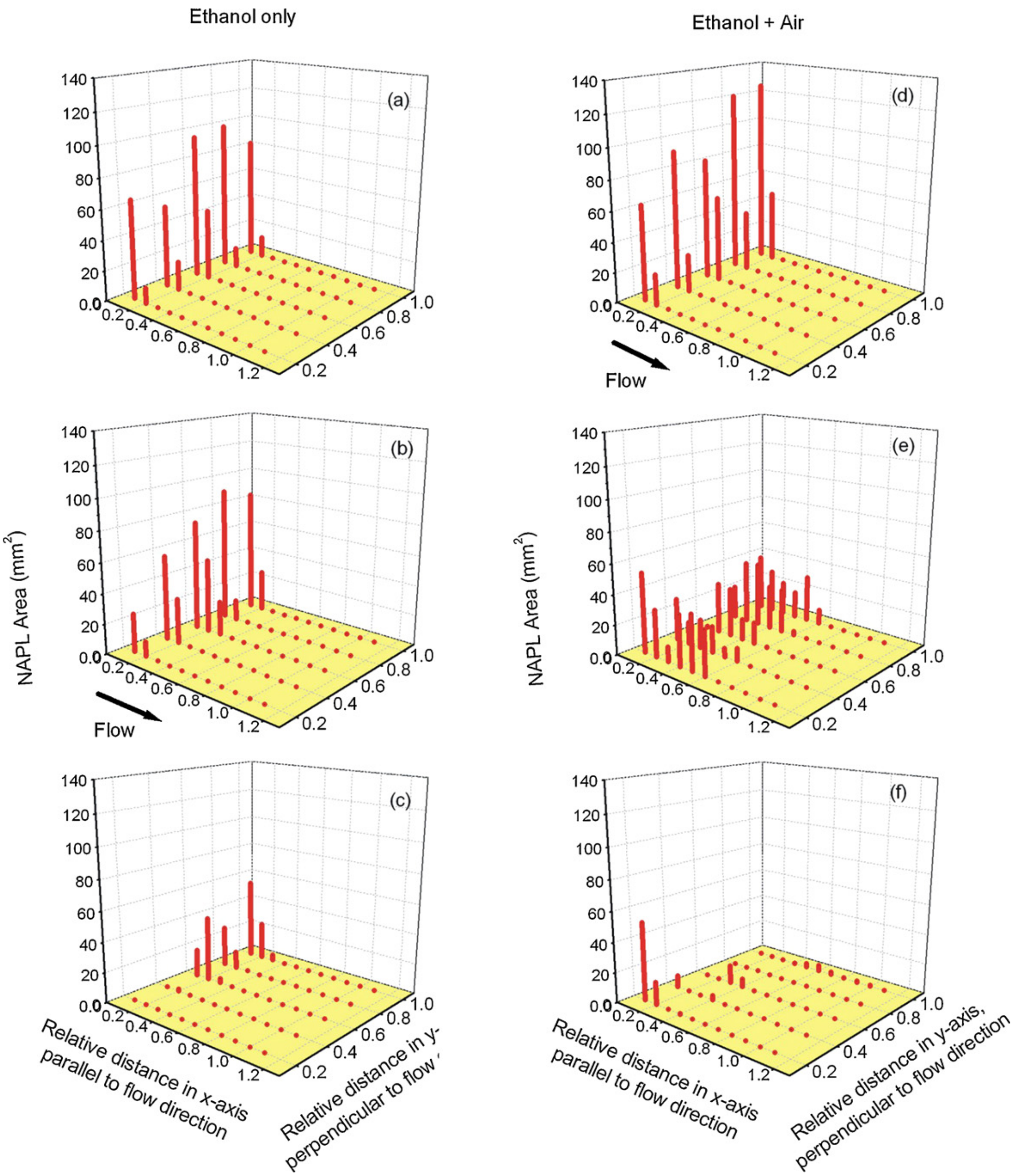

Fig. 13. Change in the benzene NAPL distribution on the two-dimensional porous medium under 70% ethanol flow conditions: (a) initial NAPL distribution; (b) NAPL distribution after 4 h elapsed under 70% ethanol flow; (c) NAPL distribution after 10 h elapsed under 70% ethanol flow; (d) initial NAPL distribution; (e) NAPL distribution after 4 h elapsed under concurrent flow 70% ethanol and air; (f) NAPL distribution after 10 h elapsed under concurrent flow 70% ethanol and air.

4. Conclusions

LNAPL swelling of TBA and 1-propanol was clearly visualized and quantified directly from the transparent porous medium. TBA and 1-propanol floods increased the benzene area, while a 70% ethanol flood only reduced the benzene area. NAPL (i.e., both DNAPL and LNAPL) swelling by cosolvent were related to alcohol partitioning to NAPL.

Partitioning types of alcohol affected dissolution of LNAPL entrapped in porous media and determined a level of the effluent concentration discharged from the LNAPL zone. The partitioning behavior of the swelling alcohols resulted in less dissolution of LNAPL.

Airflow enhanced the mobilization of LNAPL for swelling alcohol floods (TBA and 1-propanol) and the dissolution of LNAPL for ethanol flood. Therefore, NAPL behavior during cosolvent flooding would be determined by partitioning types of alcohols and the presence of airflow.

Applications of NAPL-swelling alcohols to filed remediation expect less dissolution of NAPL and increase in NAPL volume. A high flood velocity may be required for mobilization of NAPL. Mobilization of NAPL may be enhanced by providing airflow to those alcohol floods. Ethanol is expected to successfully use for the LNAPL remediation through dissolution processes. Enhanced dissolution of LNAPL would be achieved by providing airflow to ethanol flood.

Acknowledgement

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