

## EFFECT OF SOIL TEXTURE ON SURFACTANT-BASED REMEDIATION OF HYDROPHOBIC ORGANIC-CONTAMINATED SOIL

Dal-Heui Lee, Robert D. Cody\*, Dong-Ju Kim

*Department of Earth and Environmental Sciences, Korea University, Sungbuk-ku, Seoul 136-701,*

*\*Department of Water Resources, Iowa State University, Ames, IA 50011, USA*

(E-mail)

dalheui@korea.ac.kr

### 요 약 문

본 연구는 소수성 유기물로 오염된 토양의 계면 활성제를 이용한 복원에 토양의 조성이 미치는 영향에 대한 것으로서, 토양의 조성에 따른 배치실험 및 주상실험을 실시하였다. 복원 정도는 clay 함량에 따라 매우 달랐으며, clay 함량이 34% 미만일 때 복원율이 높았음을 보여주었다. 또한 clay 함량이 68% 이상일 때에는 복원율이 매우 낮았다.

**key word:** Surfactant, Remediation, Contaminant, Soil texture

### 1. Introduction

Hydrophobic organic compounds frequently enter the subsurface as a result of industrial accidents, and these chemicals represent a long-term source of soil and aquifer contamination. Their removal from contaminated soils and aquifers is difficult because they possess low solubilities and high interfacial tensions. The high interfacial tension results in large capillary forces and large displacement entry pressures that resist flushing by water. Currently the common method for remediation of aquifers contaminated with hydrophobic organic substances is pump-and-treat. This method is neither effective nor economical. However, extensive research on soil and groundwater remediation has demonstrated that surfactant-based remediation is a viable alternative for improving the efficiency of pump-and-treat remediation (Shiau et al., 1995). These studies showed that aqueous surfactant solution significantly enhanced the removal of hydrophobic organic contaminants from soil and groundwater. Even though surfactants can be used to enhance the extractive power of water, field test showed ineffective removal of hydrophobic organic contaminants where low hydraulic conductivity beds are present in the aquifer. Also, the effectiveness of surfactant-based remediation can be limited by adsorption of surfactants to clay, silt, and by organic matter (Lee et al., 2001). The objective of this study was to examine the effect of soil texture on hydrophobic organic (toluene, and 1,2,4-trichlorobenzene) removal from four kinds of soil and to evaluate the optimal composition of soil texture for maximum hydrophobic organic removal using aqueous surfactant solution.

## 2. Materials and Methods

### Surfactant selection

Four %(v/v) sodium diphenyl oxide disulfonate (DOSL,  $C_{16}H_{33}C_{12}H_7O(SO_3Na)_2$ ) anionic surfactant was selected in this study because DOSL surfactant showed a good solubilization for hydrophobic organic compounds (Chen and Knox, 1997). Rapidly biodegradable DOSL meets criteria as an indirect food additive under FDA (Food and Drug Administration of USA) Regulation (21CFR178.3400). The structure of DOSL is straight chain diphenyl oxide disulfonates. Molecular weight of DOSL is 642 and its critical micelle concentration (CMC) is 0.5 mmol/L. Also, four %(w/v) sodium lauryl sulfate (LS,  $CH_3(CH_2)_{11}SO_4Na$ ) anionic surfactant was selected for this study. LS is a food grade and also easily biodegradable. Molecular weight of LS is 288 and its CMC is 8 mmol/L.

### Soil selection

Ottawa quartz sand (U.S. Silica Company, Ottawa, IL) was chosen as a model porous medium because it has high hydraulic conductivity when packed in columns, low cation exchange capacity, and low organic carbon content. Its mean grain diameter is 0.45 mm, and specific surface area is 0.007 m<sup>2</sup>/g (U.S. Silica Company). Two Iowa soils, Fruitfield and Webster, also were selected for this study (Gonzalez and Ukrainczyk, 1996). Fruitfield soil was a sand consisting of 86.3 % (w/w) sand, 10 % (w/w) silt, 3.7 % (w/w) clay. The Fruitfield soil contained 0.2 % (w/w) organic C, its surface area was 15 (m<sup>2</sup>/g), and its pH in water was 6.2. Webster soil was a clay loam consisting of 22.9 % (w/w) sand, 43 % (w/w) silt, 34.1 % (w/w) clay. The Webster soil contained 0.61 % (w/w) organic C, its surface area was 81.4 (m<sup>2</sup>/g), and its pH in water was 8.2. One Brazilian soil was chosen as a model clay soil (Gonzalez and Ukrainczyk, 1996). Brazilian soil was a clay consisting of 6.3 % (w/w) sand, 30.4 % (w/w) silt, 64.3 % (w/w) clay. The Brazilian soil contained 0.94 % (w/w) organic C, its surface area was 43.9 (m<sup>2</sup>/g), and its pH in water was 4.8.

### Batch tests based on soil texture

Eighty grams of soil spiked with 5 mL of toluene or 1,2,4-trichlorobenzene were placed in 500 mL Teflon screw cap bottles together with 250 mL of 4 %(v/v) aqueous surfactant solution. The bottles were shaken at 200 rpm for 50 min and then centrifuged at 2000 rpm for 5 min. After decanting the supernatant, 250 mL of deionized water were added to the soil, and the bottles were shaken again for 50 min. This process was repeated until one surfctant wash and two deionized water washes were completed. Liquids were collected after each step for analysis of toluene or 1,2,4-trichlorobenzene concentration.

### Sequential column leaching tests based on soil texture

Column tests were conducted using standard methods in the organic chemistry laboratory and the soil physics laboratory of Iowa State University, USA (Lee, 1999). A porous ceramic plate beneath the soil prevented loss of soil during leaching. Briefly, 350g of soil were incremently packed to a height of 15.4 cm in glass columns (5 cm

O.D., 30 cm high). Compaction of the dry soil in 0.5 cm layers was standardized by tapping the side of the column 25 times; this degree of compaction minimizes preferential liquid channeling (Martel and Gelinas, 1996). The soil column length was 15 cm, soil column radius was 2.3 cm. When packed in columns, the soil had a bulk density of 1.40 g/cm<sup>3</sup> corresponding to a porosity of 0.47. Deionized water was pumped into the columns bottom at a rate of 3 mL/min for three hours to saturate the soil. Five mL of either toluene or 1,2,4-trichlorobenzene were injected by syringe into the middle of the column, a method of contamination analogous to a point-source of contamination at the field scale. Then 4 % (v/v) aqueous surfactant solutions was pumped into the column at a rate of 3 mL/min. Effluent was collected at each 250 mL interval for a total of fifteen intervals (3750 mL).

### 3. Results and Discussion

The greatest recovery of toluene, and 1,2,4-trichlorobenzene in batch tests was 65 %, and 75 % respectively. Contaminant removal by single head surfactant (LS) was less effective than that by the double head surfactant (DOSL) alone even though more effective than that by water (Lee et al., 2001). This is consistent with Shiau et al. (1995) who showed that twin-head anionic surfactant exhibited a greater potential than single head surfactant for solubilization of hydrophobic organic substances. This difference removal efficiency may cause from different hydrophobicity of these organic compounds. The trend of toluene and 1,2,4-trichlorobenzene removal efficiency was exactly same order with clay content of soil in all case of column test. Also, the contaminant removal efficiency of DOSL was more larger than that LS as same batch tests. Effectiveness of DOSL may be due to small adsorption of the double head sulfate polar heads onto soil particles, or to very small amounts of surfactant loss by precipitation with soil components (Lee et al., 2001).

The toluene removal of 95 % has been achieved in the Ottawa sand, and Fruitfield sandy soil after effluent volume of 3750 mL (about 32 pore volume) passed. There were little difference in toluene removal efficiency between clay loam and Brazilian soil after effluent volume of 3750 mL. A sharp increase in contaminant removal rate was observed before effluent volume of 1500 mL, and then the removal rate was very slowly increased. The contaminant removal rate was the smallest at the surfactant solution passed between 1250 mL and 2500 mL, and increased again after effluent volume of 2750 mL as the hydraulic conductivity recovered. These differences in removal rate can be attributed to the variation in soil texture, hydraulic conductivity as a function of pore volume passed. 1,2,4-trichlorobenzene removal of 98 % has been achieved in the Ottawa sand, and Fruitfield sandy soil after effluent volume of 2500 mL (about 21 pore volume) passed. Even neglecting permeability effects, these results show that sandy soils are more suitable for surfactant remediation than clay soils because of clay sorption reduces surfactant effectiveness, even though DOSL is less sorbed to clay than many other surfactants.

Surfactant solution introduced into saturated porous media can alter conductivity values by changing fluid density and viscosity values. The loss in

capability of a soil to transmit flow will decrease the efficiency of surfactant enhanced in situ remediation. Clay rich soils are less suitable candidates for surfactant-based remediation because of the effects of surfactant solutions on hydraulic conductivity. Surfactant-clay interactions which affect intrinsic permeability will also affect hydraulic conductivity and any reduction in the capability of a soil to transmit fluid will decrease the efficiency of surfactant enhanced in-situ remediation.

#### 4. Conclusion

The trend of toluene and 1,2,4-trichlorobenzene removal efficiency was exactly same order with clay content of soil in all case of batch and column tests. The toluene removal of 95 % has been achieved in the Ottawa sand, Fruitfield sandy soil after effluent volume of 3750 mL (about 32 pore volumes) passed. 1,2,4-trichlorobenzene removal of 98 % has been achieved in the Ottawa sand, and Fruitfield sandy soil after effluent volume of 2500 mL (about 21 pore volumes) passed. This result suggest that contaminated soil texture (clay content) should be considered in surfactant-assisted remediation. Also, these results show that sandy soils are more suitable for surfactant remediation than clay soils because of clay sorption reduces surfactant effectiveness, even though DOSL is less sorbed to clay than many other surfactants.

#### 5. References

- Chen, L.; Knox, R.C. Using vertical circulation wells for partitioning tracer tests and remediation of DNAPLs. *Groundwater Monitoring & Remediation*. 17: 161-168; 1997.
- Gonzalez, J.M.; Ukrainczyk, L. Adsorption and desorption of nicosulfuron in soils. *Environmental Quality*. 25: 1186-1192; 1996.
- Lee, D-H. Experimental investigation of the removal of hydrophobic organic compounds from two Iowa soils using food grade surfactants and recovering of used surfactants. Ph.D. Dissertation. Iowa State University, Ames, IA. 200pp; 1999.
- Lee, D-H.; Cody, R.D.; Hoyle, B.L. Laboratory evaluation of the use of surfactants for ground water remediation and the potential for recycling them. *Groundwater Monitoring and Remediation*. 21: 49-57; 2001.
- Martel, R.; Gelinat P.J. Surfactant solutions developed for NAPL recovery in contaminated aquifers. *Ground Water*. 34: 143-154; 1996.
- Shiau, B.J.; Sabatini, D.A.; Harwell, J.H. Properties of food grade (edible) surfactants affecting subsurface remediation of chlorinated solvents. *Environmental Science and Technology*. 29: 2929-2935; 1995.