



Laboratory evaluation of the use of solvent extraction for separation of hydrophobic organic contaminants from surfactant solutions during surfactant-enhanced aquifer remediation



Yong S. Zhao, Lu L. Li, Yan Su, Chuan Y. Qin*

Key Laboratory of Groundwater Resources and Environment, Ministry of Education, Jilin University, Changchun, PR China

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ABSTRACT

Solvent extraction is an efficient method that separates surfactants and pollutants in pumped groundwater during surfactant-enhanced aquifer remediation (SEAR). This paper addresses the factors that influence the extraction efficiency of solvent extraction method. A series of batch experiments were conducted to verify the effectiveness of solvent extraction by describing the interactions among contaminants, surfactants, and solvents, using factors such as extraction time, solvent/water volumetric ratio, surfactant type and concentration, contaminants type and concentration, solution salinity and solvent equivalent alkyl carbon numbers (EACNs). The surfactants, contaminants, and solvents used were sodium dodecyl sulfate (SDS) and Tween 80, benzene and nitrobenzene, *n*-hexane, *n*-decane and *n*-hexadecane, respectively. The results indicated that (1) the increase in extraction time or solvent/water volumetric ratio caused higher benzene removal efficiency, but the increment of removal was not significant after 2 h or 0.1; (2) surfactant type and concentration significantly affected extraction efficiency. The increase in SDS concentrations could generally increase benzene removal and then decline, whereas benzene removal seldom changed under the studied Tween 80 concentration range. Moreover, Tween 80 losses were much lower than that of SDS. When mixing the two surfactants, benzene was increasingly separated from the aqueous surfactant solutions by increasing the proportion of Tween 80, and surfactant losses almost depended on monomers instead of micelles within the aqueous surfactant solutions; (3) the efficiency of separating benzene from aqueous surfactant solutions was higher than that of nitrobenzene. Furthermore, *n*-hexane losses in the benzene aqueous solution were higher than that in the nitrobenzene aqueous solution; (4) higher inorganic salt concentration in the aqueous Tween 80 solutions could increase benzene removal and decrease Tween 80 losses, thereby improving extraction efficiency; (5) *n*-hexane was more suitable to separate benzene from aqueous Tween 80 solutions because its EACN was closest to the EACN of benzene.

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1. Introduction

The contamination of soils and groundwater by non-aqueous phase liquids (NAPLs) is a major environmental problem worldwide [1–3]. Although not applicable to every situation, surfactant-enhanced aquifer remediation (SEAR) proved a promising technology to clean NAPL contaminants, such as benzene and nitrobenzene from contaminated soils and groundwater [4–7]. Compared with traditional pump-and-treat remediation, SEAR could overcome the “tailing effect”, and thus remediation time could be dramatically reduced, thereby improving remediation

efficiency. Therefore, SEAR has been applied widely to numerous fields contaminated by organic compounds in soils and groundwater. Researchers worldwide, particularly in the United States, have increasingly studied this technology. The key to the effective use of SEAR is the selection of surfactants. Surfactants are amphiphilic, surface-active molecules that contain both hydrophilic and hydrophobic groups [8,9]. Upon a certain concentration, known as critical micelle concentration (CMC), the surfactants would form spherical or cylindrical micelles with their hydrophilic head groups shielded with organic pollutants and the hydrophobic tail groups toward the aqueous phase [10]. Following the nature of the hydrophilic portion of the molecules, the surfactants are typically classified into cationic, anionic, zwitterionic, and nonionic surfactants [9]. Anionic and nonionic surfactants are often considered for SEAR to minimize surfactant sorption losses.

* Corresponding author. Tel.: +86 431 88502608.

E-mail address: qincyu@jlu.edu.cn (C.Y. Qin).

Although considerable progress has been made regarding remediation performance with SEAR compared with pump-and-treat [11–13], remediation may cause a substantially adverse environmental impact because of the intensive usage of surfactants [14–16]. Treatment efficiency may not be desirable in certain engineering applications because of abundant foam generated during wastewater processing. Moreover, SEAR is not economically feasible without a means for surfactant recovery and recycling [17]. High treatment efficiency and significant cost savings of SEAR can be achieved by reusing surfactants after separating the organic compounds from the surfactant solutions. In the last two decades, with considerable advances in SEAR research and implementation, additional efforts have focused on the removal of organic compounds from aqueous surfactant solutions. At present, efficient separation methods are widely used to partition organics and surfactants, e.g., pervaporation, air stripping, foam separation, reverse-micellar extraction, vacuum extraction, and solvent extraction [11,18,19].

Pervaporation is a membrane-based process to separate liquid mixtures [20]. Laboratory investigations have been conducted to separate volatile organic compounds (VOCs) from waste solution of surfactants and surfactant-based solvents [11,18,21]. Pervaporation system performance factors were also studied [22]. Foam fractionation could be another alternative to move VOCs. The recovery of surfactants from water by multistage and simple continuous mode foam fractionation were attempted for the applicability of foam separation [14,16]. However, significant surfactant foaming could occur and its equipment cost is substantial. These technologies, as well as air stripping, were merely used to remove VOCs from surfactant solutions [23]. Solvent extraction has been tested as a well-established technology to separate organic compounds from pumped groundwater in SEAR for its high stability and efficiency. The approach introduces a solvent to the surfactant solution such that the organic compounds would transfer from the surfactant solutions to the solvent. The extraction efficiency largely depends on the interfacial area, which is generated by the mixing of two phases, namely aqueous phase and solvent phase. Fig. 1 shows the distribution of surfactant and contaminant molecules in a solvent extraction system [4]. A series of experiments were conducted to study the performance of the method and to examine the effects of various parameters on the separation efficiency of the

surfactants and contaminants. The parameters include the type of extracting solvents, concentration of surfactants, type of contaminants, solvent/solution volumetric ratio and inorganic salts [4,24–28]. Cheng et al. [4] studied solvent extraction to remove contaminants with low equivalent alkyl carbon numbers (EACNs) from surfactant solutions, with surfactant concentration, solution salinity, and solvent/solution volumetric ratio as the influencing factors. The results showed that extracting solvents must have much higher EACNs than that of the contaminant. However, the highest EACN solvent was not necessarily optimal for contaminant removal because of other constraints. Moreover, increasing the total surfactant concentration or salinity of an anionic surfactant solution could increase its contaminant solubilization capacity, but reduce the contaminant removal efficiency. Montilla et al. [24] investigated the removal of pollutants from water with a surfactant, solvent, or alcohol via two solvent extraction methods, with polar and non-polar contaminants as the controlling factors. Both methods have been shown to be effective and efficient to remove the hazardous pollutants from an aqueous solution. Lee et al. [25] used a glass column to determine the efficiency of solvent extraction for used surfactant recycling, with acetone, hexane, and methylene chloride as the solvents and toluene and 1,2,4-TCB as the pollutants. The results showed that toluene and 1,2,4-TCB were effectively removed from the aqueous surfactant solutions. The time required to remove 98% of toluene and 1,2,4-TCB was 5 h with solvent flow rate of 30 mL/min. This result indicated that solvent extraction was an excellent method to recycle surfactant solutions during SEAR.

The majority of prior studies considered contaminant removal as the major evaluation of extraction efficiency, however, relevant contemporary studies that fully address the interactions among the contaminant, surfactant, and extracting solvent, especially surfactant and extracting solvent losses are scarce. Therefore, a series of batch experiments were conducted in this paper to verify the effectiveness of solvent extraction by quantifying the interactions among contaminants, surfactants, and solvents, with limitation factors, such as extraction time, solvent/water volumetric ratio, surfactant type and concentration, contaminants type and concentration, solution salinity and solvent EACN. The major objectives of this study are (1) to investigate the influence of extraction time and solvent/water volumetric ratio on contaminant removal

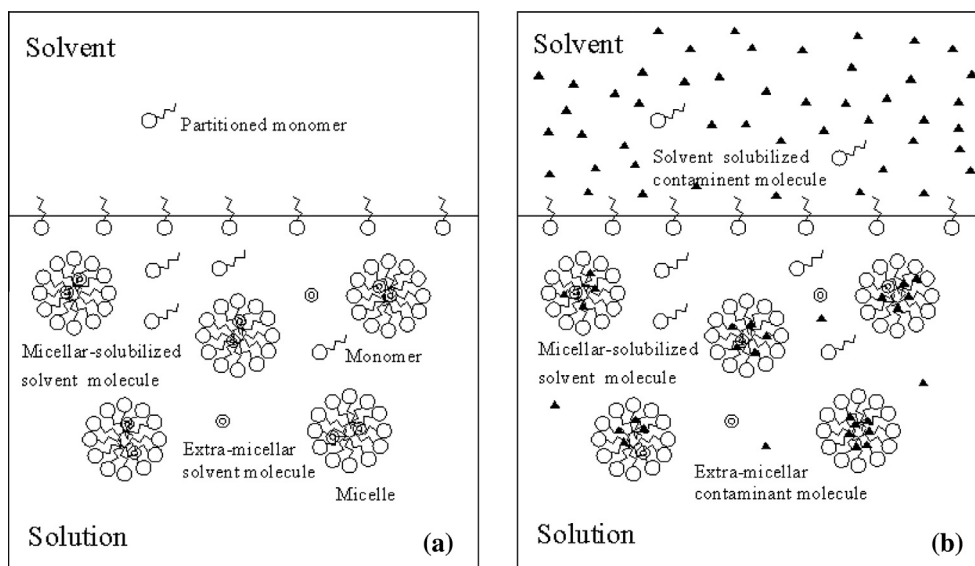


Fig. 1. Interactions among surfactant, contaminant, and extracting solvent and corresponding species distributions in solvent extraction system: (a) surfactant and solvent interactions and (b) surfactant, contaminant and extracting solvent interactions [4].

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