



Adsorption of mixed polycyclic aromatic hydrocarbons in surfactant solutions by activated carbon



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ABSTRACT

The adsorption behavior of three polycyclic aromatic hydrocarbons (PAH) in TX100 solution by activated carbon was studied aiming at surfactant recovery. Adsorption experiments were conducted in batch and column models. The adsorption data fitted well with the Langmuir isotherm model, Dubinin–Radushkevich isotherm models and pseudo-second-order kinetics model. Regarding column adsorption, the exhausting time of TX100 was shorter than the breakthrough time of PAH in the fixed AC column. This finding indicates the feasibility of reusing surfactants from soil-washing solutions. The total costs can reduce about \$ 0.57 per 10 L washing solution with AC adsorption.

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1. Introduction

The remediation of hydrophobic organic compounds (HOCs), such as polycyclic aromatic hydrocarbons (PAHs), is a challenging problem [1,2]. PAH contamination usually manifests as a complex mixture of compounds resulting from both the combustion of carbon-containing fuels and the accidental or improper disposal of industrial materials [3–5]. PAHs are typical hydrophobic organic molecules that contain one or more benzene rings arranged in various configurations. They are found in high concentrations especially in many industrial sites and urban soils [6]. PAHs are relatively stable and less bioavailable for microbial degradation than many other organic compounds [7,8]. Therefore, PAHs are also classified as persistent organic pollutants because they tend to remain in the environment for a long time.

Surfactant-enhanced aquifer remediation (SEAR) has been proposed as a promising remediation technology because of its relatively high removal ratio and cost effectiveness [9–11]. SEAR is implemented by injecting dilute aqueous surfactant solutions into contaminated soil. Surfactant solutions can lower the interfacial tension between organic and aqueous phases, as well as increase the apparent solubility of HOCs via micelle solubilization above its critical micelle concentration (CMC) [12–14]. The most serious impediment of the full-scale application of the technology is the

cost of the surfactant due to surfactant cost can increase operational expenses by up to 50% [15]. Separation of organic contaminants from surfactant solutions is necessary to reuse surfactant solutions and to lessen the demand on waste disposal. Many processes can be employed to remove HOCs from contaminated surfactant solutions based on different principles. Separation processes through adsorption have the advantages of low operational and maintenance cost, low pollution, and few equipment investments. Given the high production cost of activated carbon (AC), AC should be regenerated or reused. Adsorption technology can be utilized as a surfactant recovery method in surfactant-enhanced remediation.

However, studies on the reuse of surfactant solutions by AC are rare. It was reported that adsorption is not practical for the treatment of high concentrations of waste for surfactant reuse by AC [16]. However, Ann proposed that adsorption by AC is an ideal alternative for recovering surfactants because the partitioning coefficient of phenanthrene (PHE) is higher than that of nonionic surfactants [17–19]. Wan also verified the capability of activated carbon on selective removal of contaminant while the surfactant was recovered and reused [20].

Considering that contaminants naturally exist as a mixture, the adsorption characteristics of mixed contaminants may be different from those of single contaminants in surfactant solutions. The present study aims to investigate the performance of AC in the selective adsorption of HOCs from nonionic surfactants and to address the problems associated with multicomponent adsorption from surfactant solutions. A mixture of three PAHs often found

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together in soil-washing solutions was studied, namely, PHE, fluoranthene (FLA), and benzo(a)anthracene (BaA). TritonX-100 (TX100) was used as a nonionic surfactant because target pollutants can be removed at a larger amount and fewer losses are incurred by sorption onto the soil [21–25]. Adsorption experiments were carried out both in batch and fixed-bed column systems. The parameters of the adsorption isotherm and kinetics models were determined. This work was supposed to provide further information on the applicability of surfactant reuse by AC adsorption.

2. Materials and methods

2.1. Materials

The nonionic surfactant TX100 with purity >98% was purchased from J&K Scientific Ltd. TX100 is a nonionic surfactant with a molecular weight of 625 g/mol and CMC of 150 mg/L. Three polycyclic aromatic hydrocarbons, PHE, FLA, and BaA (Tkyo chemical industry Co. Ltd., purity >97%) were employed in the studies. Selected physical and chemical properties of these PAHs are provided in Table 1. All the above chemicals were used without further purification.

Commercial AC supplied by Sinoharm Chemical Reagent Co., Ltd., China, was used as the adsorbent. The AC was boiled in deionized water for 1 h, purged with deionized water for 3–5 times, then dried under 105 °C over night and stored in a dryer before use. The pore structure characteristics of AC were determined by nitrogen adsorption at 77 K using ASAP instrument. The Brunauer–Emmett–Teller (BET) surface area and pore size distribution were obtained by BET equation and density function theory method. Carbon (C), hydrogen (H), and nitrogen (N) contents of the adsorbents were determined by an element analyzer and oxygen (O) content was calculated by difference.

2.2. Solubilization experiments

For each batch test, excess amount (about four times of the apparent solubility) of an individual PAH was added to each glass tubes containing a series of 25 mL TX100 surfactant solutions with various concentrations to ensure maximum solubility. These samples were then equilibrated for a period of 48 h on a reciprocating shaker at approximately 200 rpm maintained at a temperature of 25 ± 0.5 °C. After shaking, the vials were allowed to settle for at least 15 h then subsequently

centrifuged at 4000 rpm for 20 min to separate the undissolved PAHs. The solubilization experiments were duplicated and the average values were calculated.

2.3. Batch adsorption experiment

A stock solution was prepared by dissolving individual or mixed PAHs in TX100 solution. The concentration of PHE, FLA, BaA, and TX100 is 120, 80, 12, and 5000 mg/L respectively in stock solutions. This was diluted to obtain the required concentrations for further use. The activated carbons (1 g/L) were added to 150 mL glass flasks and then filled with 100 mL solution containing both TX100 and individual or mixed PAHs. The adsorption equilibrium isotherms for individual or mixed PAHs were carried out on a reciprocating shaker at approximately 200 rpm maintained at a temperature of 25 ± 0.5 °C for 24 h. Blank samples (without carbon) were also used to determine the value of PAH or TX100 volatilization and adsorption on the surface of the glass flask. The blank recoveries ranged from 94% to 98% and the data were adjusted for these recoveries. In adsorption kinetics experiment, the aqueous samples were taken at 10, 20, 30, 40, 60, 120, 180, 240, 480, 600, 900, 1080, and 1440 min. After shaking, the suspension was centrifuged at 5000 rpm for 10 min.

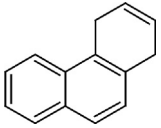
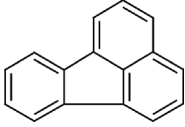
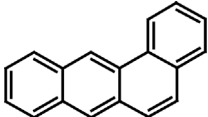
2.4. Fixed bed adsorption experiment

The fixed bed experiment was conducted using organic glass column of 2.5 cm inner diameter and 20 cm height. The column was flushed with de-ionized water for 1 h and dried at 80 °C for 24 h, then filled with activated carbon. Solution was prepared by dissolving PAHs at a known mass in TX100 solution. The various parameters like bed depths (4.7, 10.8, and 14.7 cm), flow rates (10, 20, and 30 mL/min), influent TX100 concentration (3, 4, and 5 g/L), and influent PAH concentration (PHE concentration is 60, 90, and 120 mg/L, respectively) were taken in the fixed bed experiment. The solutions at the outlet of the column were collected at every 15 min.

2.5. Regeneration of the saturated activated carbon

After adsorption, AC was dried at 105 °C for 6 h and regenerated in a quartz reactor by 2500 MHz microwave irradiation at 800 W for 6 min. The experimental setup have been described elsewhere [29]. The regeneration activated carbon was reused for further cycles of adsorption in a batch study.

Table 1
Selected properties of three PAHs.

PAHs	MS [26]	MW (g/mol) [26]	MD (Å × Å × Å) [26]	Cs 25 °C (μg/L) [27]	Log K _{ow} [28]
Phenanthrene PHE		178.2	11.7 × 8.0 × 3.4	1290	4.57
Fluoranthene FLA		202.26	11.4 × 9.5 × 3.8	200–260	5.22
Benzo(a)anthracene BaA		228.3	13.9 × 3.7 × 3.9	10	5.91

MS: molecular structure; MW: molecular weight; MD: molecular dimension; Cs: water solubility; K_{ow}: octanol–water partition coefficient.

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