



Enhanced solubilization and desorption of pyrene from soils by saline anionic–nonionic surfactant systems



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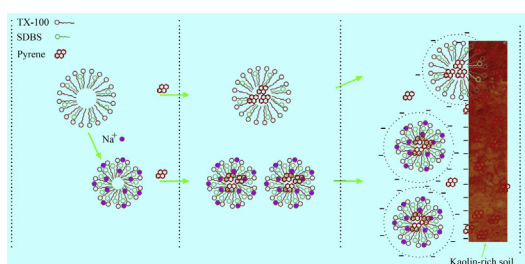
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HIGHLIGHTS

- Saline surfactant shows lower *CMC* values than fresh surfactant.
- The lower β values of saline surfactants, the higher pyrene solubilization.
- STPP reduces surfactant sorption onto soil and enhances pyrene desorption.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigates the influence of salinity on micellar behavior, enhanced solubilization and desorption of pyrene within single and mixed saline anionic–nonionic surfactants. Various interaction parameters linked to the micellization and solubilization process have been correlated through theoretical treatments to quantify micellar characteristics and solubilization capabilities of the systems. Results show that the experimental critical micelle concentration (*CMC*) values of the saline system were lower than that of non-saline system, indicating more attractive interactions of individual surfactants and higher stability of mixed micelles in the presence of salts. Pyrene solubility in saline surfactant solutions exceeded that in a non-saline solution. Less electrostatic repulsion of saline mixed micelles may be responsible for the reduced *CMC* of the mixed micelles, the increase of molar solubilization ratio (*MSR*) and micelle–water partition coefficient (K_m). The saline micelles enhanced desorption of pyrene from kaolin-rich soil, and the pyrene desorption increased with the increase of salinity. Furthermore, the presence of salinity reduced the sorption of surfactant onto kaolin-rich soil; e.g. from 15.34 to 11.07 mg of surfactant/g of soil at a mass ratio of 5:5 mixture of anionic and nonionic surfactants. The results of this study provide new insights for estimating the utility of mixed surfactants treatments for soil remediation.

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1. Introduction

The contamination of soil, sediment and groundwater by polycyclic aromatic hydrocarbons (PAHs) is a widespread concern due to the toxic risks of PAHs to the environment and public health [1]. PAHs are formed during natural and anthropogenic pyrolysis of organic matter [2–4], and anthropogenic emissions of PAHs

are increasing, particularly in developing countries (e.g. China) [5], leading to widespread pollution of the environment, including soils.

Like many hydrophobic organic compounds, PAHs can strongly sorb to soils, especially to the soil's clay fraction [6,7], enabling them to persist in soils for long periods of time. Various attempts have been made to remove PAHs from contaminated industrial sites. Surfactant enhanced remediation (SER) has been proposed as a promising option for treating PAHs-contaminated soils [8–10]. A surfactant consists of both a hydrophilic polar head group and a hydrophobic hydrocarbon chain, which can form micelles in the aqueous phase when the concentration of surfactant exceeds the critical micelle concentration (CMC) [11]. These micelles can enhance the solubilization of PAHs and their transfer from contaminated soil to the aqueous phase [3].

In some cases, a single surfactant can sorb on the surface of the soils, inevitably lowering its ability to desorb PAHs. To address this limitation, a mixture of anionic–nonionic surfactants has been investigated in a number of scientific studies [12,13]. Because adding nonionic surfactants to anionic surfactant solutions can decrease the interfacial force, result in a more compact molecular arrangement and reduce precipitation that can occur between anionic surfactants and divalent electrolytes; e.g. Ca^{2+} , Mg^{2+} [9,13,14]. Also, mixing an anionic surfactant with a non-ionic surfactant can reduce the electrostatic attraction and inhibit the sorption of non-ionic surfactants to soil surfaces [15]. As a result, in practical applications researchers attempt to use mixed surfactants to reduce surfactant losses and improve desorption efficiency of the SER for contaminated soils.

To effectively use mixed surfactants to enhance the solubilization and desorption of PAHs requires knowledge of the surfactant and target environments and an understanding of their complex interactions [16,17]. One significant environmental factor impacting the interactions is the salinity or ionic strength of the solution, which may affect both the CMC and the sorption loss of anionic surfactant [18,19]. For the nonionic surfactant, the influence of salinity on the solubilization is comparatively slight; however, adding salt to an anionic surfactant solution can decrease the CMC value [20–22]. Some researches have shown that surfactants with a higher salinity solubilize larger amounts of hydrophobic organic compounds (HOCs) due to reduced CMC values and increased micelle numbers, which facilitates the mobility of HOCs, leading to improved soil washing [23]. However, other researches have shown that increasing salinity decreases solubility of HOCs and spreads the distribution of HOCs to soil phases [24,25] due to interactions between the surfactant and salt that makes it difficult for HOCs to incorporate into the hydrophobic core of micelles [18,26]. In effect, the presence of more salt ions makes HOCs more persistent in soil particles and adversely influences their desorption from the soils [3,27]. On the other hand, the effect of salinity on solubilization and desorption of PAHs by anionic–nonionic mixed surfactants is less clear, as is its effect on micellar behavior.

The objectives of the present work are to study the effect of salinity on CMC, solubilization and desorption of PAHs using mixed micelles. Based on our CMC values of single and mixed surfactants and aqueous solubility data, models proposed by Clint [28], Rubingh [29] and Attwood [30] have been used to explore possible

synergism in the solubilization process. As a practical application, the performance of single and mixed anionic–nonionic surfactants and sorption of surfactants onto kaolin-rich soils in Guangdong, China was also studied. The experimental results of this study are useful in understanding the role of salinity in SER.

2. Methods

2.1. Materials

Pyrene was selected as a representative PAH and obtained from Aldrich Chemical Company with a purity greater than 98%. The nonionic surfactant *t*-octylphenoxypolyethoxyethanol (TX-100) and anionic surfactant sodium dodecyl benzene sulfonate (SDBS), each with a purity of 99.9%, were obtained from Sigma Chemical Company. Both pyrene and surfactant are used as received. The properties and molecular structures of pyrene, TX-100 and SDBS are given in Table S1 and Fig. S1 respectively (Supplementary Material). Reagent grade sodium chloride, sodium tripolyphosphate (STPP) and sodium carbonate were obtained from Tianjin Kemiou Chemical Company. Acetone was purchased as analytical grade solvent. Deionized water was used throughout the study.

2.2. Determination of CMC

The CMC values of the pure and mixed TX-100 and SDBS solution were determined in the presence and absence of salinity (1.8% (m/m) sodium chloride, STPP and sodium carbonate) by the surface tension method. The mass ratios of TX-100 and SDBS were set at 10:0, 9:1, 7:3, 5:5, 3:7, 1:9 and 0:10. Surface tension of solutions was tested at 25 °C by the Wilhelmy plate technique with a QZBY-1 model tensiometer (Fangrui Instrument Co., Ltd., Shanghai). The surface tension value was taken when a stable reading was obtained at a given surfactant concentration, as indicated by at least three consecutive measurements displaying the same values. The CMC values were determined by plotting the surface tension versus the surfactant concentration [5].

2.3. Preparation of pyrene contaminated soils

Soil samples were taken from Guangzhou University Town, China, collected at a depth of 200 mm below the surface. Physicochemical properties of these soils are listed in Table 1. The soils were air-dried at ambient temperature (25–30 °C) for one week and passed through a 2 mm sieve to remove coarse fragments. Contaminated soils were then prepared by dissolving pyrene in acetone and mixing the liquid into the soils for 30 min to obtain soils contaminated with 100 mg of pyrene/kg of soil. The samples were placed in a hood, uncovered, for 24 h and aged for 7 days in a closed vessel at room temperature.

2.4. Preparation of single and mixed micelles and solubilization tests

Appropriate amounts of pyrene and 20 mL mixed surfactant solution were added to a series of 30 mL borosilicate vials in the

Table 1
Properties and clay mineral compositions of the selected soil.

BET ^a (m ² /g)	CEC ^b (mmol/kg)	Organic matter ^b (%)	pH ^b	Total clay contents (%)	K ^c (%)	V ^c (%)	I/V ^c (%)
10.6	54.1	0.16	5.89	36	90	4	6

^a The BET surface area of the samples was determined by the nitrogen sorption isotherms measured at 77.3 K on a surface area analyzer (Micromeritics Tristar II 3020, USA).

^b The pH, organic matter, and cation exchange capacity (CEC) of the samples were determined by the standard methods described in Carter [31].

^c Relative contents of clay minerals. K: kaolin, V: vermiculite, I/V: illite-vermiculite mixed-layer. Total and relative contents of clay minerals were determined by XRD.

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