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Adsorption of sodium dodecyl sulfate, Triton X100 and their mixtures to shale and sandstone: A comparative study

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ABSTRACT

In this paper, batch experiments of sediment/aqueous systems were conducted to evaluate the adsorption of SDS, TX100 and their mixtures (1:2; 1:1 and 2:1 SDS:TX100 mass ratio) onto local shale and sandstone. Adsorption of surfactants was assessed using a surface tension technique for surfactant concentrations less than surfactant monomer saturation (CMC). It is shown that the amount of TX100 adsorbed to shale (7.5 g/kg) are greater than those adsorbed to sandstone (1.5 g/kg). SDS showed negligible affinity for adsorption on both adsorbents. The amounts of both TX100 adsorbed to shale or sandstone can be decreased and minimized when they are mixed with SDS. While adsorption of TX100–SDS mixtures on shale reduced to 4.5 g/kg (40% reduction in comparison to adsorption of TX100), adsorption to sandstone decreased tremendously to 0.3 g/kg (80% reduction in comparison to adsorption of TX100). Furthermore, micellization behavior was assisted through mixing. CMCs of mixtures reduced to 0.1 wt.% in presence of shale compared to 0.15 wt.% for TX100 and 0.1 wt.% for SDS. Similarly, CMC of mixtures reduced to 0.03 wt.% in presence of sandstone in comparison to 0.05 for pure TX100 and 0.1 wt.% for SDS. Because of their ability to minimize amounts adsorbed in different adsorbents, mixed anionic–nonionic surfactant particularly TX100–SDS may show potential advantages in SEAR and EOR applications.

1. Introduction

Surfactant adsorption is a process of transfer of surfactant molecules from bulk solution phase to the surface/interface. It can be considered as a partitioning of the adsorbate species between the interface and the bulk, and can occur if the interface is energetically favoured by the surfactant in comparison to the bulk solution (Paria and Khilar, 2004; Zhang and Somasundaran, 2006; Parida et al., 2006).

The adsorption of surfactants on rock/soil/sediment solid matrix may result in the loss and reduction of their concentration, which may render them less efficient or ineffective in practical treatment. Adsorption of surfactants from aqueous solutions in porous media is very important in enhanced oil recovery (EOR) of oil reservoirs because surfactant loss due to adsorption on the reservoir rocks impairs the effectiveness of the chemical solution injected to reduce the oil–water interfacial tension (IFT) and renders the process economically unfeasible (Curbelo et al., 2007). Furthermore, the adsorption of surfactants by soils can result in much surfactant being unavailable for the micellar solubilisation of hydrophobic organic compounds (HOCs), thereby decreasing its efficiency for desorption. Nevertheless, the presence of adsorbed or immobilized surfactant in soils gives rise to the partition or adsorption of HOCs in the soil–water system, which may contribute to a decrease in the desorption of organic compounds (Edwards et al., 1994; Sun et al., 1995; Haigh, 1996; Ko et al., 1998). In this sense, comparable to the selection of a surfactant to solubilise the contaminant or reduce the oil IFT is its minor tendencies to be adsorbed by the soil/sediment/reservoir rock under consideration.

The adsorption of single surfactants at the pure solid–liquid interface has been studied intensively, but far fewer studies exist for the case of adsorption of single surfactants to natural soils. Nevertheless, adsorption of surfactant mixtures to natural soils is even scarce. Generally, the literature contains several studies about the adsorption of TX100 by soils/sediments/clay minerals. Studies on the adsorption of SDS are comparatively less common and even far fewer studies deal with adsorption of TX100–SDS mixture to soils/sediments/clay minerals (Yu et al., 2007; Zhou and Zhu, 2007a).

Surfactant adsorption by soils/sediments depends on the type of surfactant and the soil properties. In this sense, the results of different studies addressing the adsorption of anionic and nonionic surfactants by soils/sediments with different compositions have revealed the relative importance of soil/sediment organic matter and clay minerals on the adsorption of anionic and nonionic surfactants.

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Nomenclature

SDS	Sodium dodecyl sulphate
TX100	Triton X-100
CMC	Critical micelle concentration
SEAR	Surfactant enhanced aquifer remediation
EOR	Enhanced oil recovery
IFT	Interfacial tension
ST	Surface tension

Urano et al. (1984), Fytianos and co-workers (Fytianos et al., 1998a, b) and Paterson et al. (1999) found the adsorption of anionic and nonionic surfactants as a function of soil/sediment organic carbon contents. However, a vast majority of researchers showed that adsorption of anionic/nonionic surfactants onto soil/sediments is governed primarily by the fraction of swelling clays (Podoll et al., 1987; Cano and Dorn, 1996a,b; Ou et al., 1996; Brownawell et al., 1997; Yuan and Jafvert, 1997; Salloum, 1999; Salloum et al., 2000; Lee et al., 2000; Zhu et al., 2003; Rodriguez-Cruz et al., 2005; Lee et al., 2005; Zhou and Zhu, 2007b; Sanchez-Martin et al., 2008).

Furthermore, Brownawell et al. (1997), Lee et al. (2000, 2005), Salloum (1999), Salloum et al. (2000) and Zhu et al. (2003) demonstrated that the adsorption of nonionic surfactant on soils/sediments may be a combination of organic matter fractions and clay mineral fractions. However, the clay mineral fractions particularly, montmorillonite and illite, have by far a stronger affinity to nonionic surfactant compared to organic matter fractions.

Zhu et al. (2003), Rodriguez-Cruz et al. (2005) and Sanchez-Martin et al. (2008) have attempted to study the influence of the mineralogical composition of the clay fraction in the adsorption of surfactants and found illite and montmorillonite have a great affinity to TX100, while kaolinite has far less affinity. Humic matter has even far less adsorption affinities. The order that adsorption capability for montmorillonite is higher than that for kaolinite is reported also by Salloum (1999) and Shen (2000).

Nonionic surfactants were found to adsorb extensively to pure clay minerals, however, adsorbed amounts of anionic surfactant to montmorillonite were significantly lower. Furthermore, adsorbed amounts of anionic/nonionic surfactants to natural soils are significantly less than those reported for pure clay minerals (Edwards et al., 1992; Zhu et al., 2003; Zhou and Zhu, 2007a,b; Yu et al., 2007).

It is clear that adsorption of nonionic surfactant, TX100, to pure clay minerals is very significant if compared to its adsorption to natural soils containing a few percentage of similar clay minerals. Noticeably, the effects of soil/sediment clay–mineral composition on adsorption of nonionic surfactants by soils/sediments are more significant than the case of anionic surfactants.

Adsorption of anionic–nonionic surfactant mixtures mainly focused on pure solids (Scamehorn et al., 1982a,b; Harwell et al., 1988; Xu et al., 1992; Somasundaran et al., 1992a; Somasundaran et al., 1992b; Ma and Xia, 1992a,b; Denoyel et al., 1993). Results showed that when single surfactant adsorption was investigated on positively charged surfaces, only the anionic surfactants were found to adsorb to any appreciable extent on rutile/alumina. In the mixed system, the anionic surfactant was found to induce significant co-adsorption of the nonionic surfactant (Ma and Xia, 1992a; Somasundaran et al., 1992a,b). Anionic surfactants, which by themselves do not adsorb on negatively charged silica/kaolinite, shows significant adsorption in the presence of nonionic surfactants and markedly changes the zeta potential of silica particles (Somasundaran et al., 1991; Somasundaran et al., 1992a,b; Denoyel et al., 1993). Similarly, on negatively charged surfaces, it was shown that the maximum adsorption of nonionic surfactant on silica decreased in presence of anionic surfactant (Gao et al., 1984). However, the adsorption of both surfactants on carbon black was found to decrease in the presence of the other component (Ma and Xia, 1992b).

More recently, adsorption of TX100–SDS surfactant mixtures on different pure materials was reported (Wang and Kwak, 1999; Somasundaran and Huang, 2000; Paria and Khilar, 2004; Yang et al., 2005). Generally, similar conclusions were reached. For the adsorption case of anionic–nonionic surfactant mixtures, which surfactant is adsorbed preferentially depends on the nature and charge of mineral surface. For positively charged surfaces to which anionic surfactants are heavily adsorbed, presence of nonionic surfactant decreases adsorption of anionic surfactant and the adsorption of nonionic surfactant is enhanced where nonionic alone shows trace adsorption (Wang and Kwak, 1999; Paria and Khilar, 2004). On the other hand, for clay minerals that show affinity to nonionic surfactant (Ca-montmorillonite), presence of anionic surfactant retards nonionic surfactant adsorption (Paria and Khilar, 2004; Yang et al., 2005).

By comparison very few studies have been reported on adsorption of anionic–nonionic surfactant in natural soils (Rao and He, 2006; Yu et al., 2007; Zhou and Zhu, 2007a). Zhou and co-workers' (Yu et al., 2007; Zhou and Zhu, 2007a) results showed that, when SDS was mixed together with TX100, the adsorption of nonionic surfactant onto soil was severely restricted and a higher mole fraction of SDS in surfactant solution meant that a lower plateau adsorption can be reached with a smaller TX100 concentration in surfactant solution. Reduction of adsorption loss in mixed surfactants indicated that mixed surfactant could increase available concentration of the surfactant quantities and then reduce the costs and the level of surfactant pollution in remediation.

Recent successes in reducing adsorption of nonionic surfactants by mixing with anionic surfactants are encouraging and warrant the use of the technique in appropriate local soils. This study is intended to investigate the simultaneous losses of SDS, TX100 and their mixtures by adsorption. The study will investigate the adsorption of SDS + TX100 to local shale and sandstone. Surface tensions of surfactants and their mixtures are studied to determine adsorption and micellization properties.

2. Materials and methods

2.1. Surfactants

Polyethylene glycol tert-octylphenyl ether-Triton X-100 (TX100) was supplied by Scharlau Chemie, Spain. It is a nonionic surfactant ($C_8H_{17}C_6H_4(OCH_2CH_2)xOH$) with a CMC which spans between 0.13 and 0.2 g/L (0.2–0.31 mmol/L) (Zhu and Feng, 2003; Zhou and Zhu, 2004; Rodriguez-Cruz et al., 2005; Yang et al., 2005; Zhao and Zhu, 2006; Yu et al., 2007; Zhou and Zhu, 2007a,b) and it has an average of 9.5 ethylene oxide units per molecule with an average molecular weight of 646.37 g/mol. Sodium dodecyl sulphate–SDS ($C_{12}H_{24}NaSO_4$) is an anionic surfactant with a CMC which spans between 1 and 2.4 g/L (3.32–8.4 mmol/L) (Zhu and Feng, 2003; Zhou and Zhu, 2004; Rodriguez-Cruz et al., 2005; Zhao et al., 2005; Yu et al., 2007; Zhou and Zhu, 2007a) and its molecular weight is 288.4 g/mol–was supplied by Merck with a high grade of purity (99%). All chemicals were used as received without further purification.

2.2. Sediment samples

Shale samples were collected from an outcrop of a local shale formation (Batu Arang, Selangor, Malaysia). Sandstone samples were collected from an outcrop of a local sandstone formation (Bukit Bangkong, Kuala Rompin, Pahang). Samples were disintegrated into small pieces by jaw crusher and then ground using rock pulverizer (Fritsch, Germany). Samples were air dried for 24 h followed by oven drying at 105 °C for 24 h. Dried rock samples were sieved