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Study on the sorption behaviors of Tween-80 on marine sediments

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ABSTRACT

Batch experiment was carried out to study the sorption behavior of polysorbate 80 (Tween-80) on marine sediments collected from three different sites in Bohai Sea, China. The sorption of Tween-80 reached equilibrium within 3 h and the sorption kinetic curve could be divided into two sections: the rapid sorption part and the slow sorption part. The initial sorption rate increased with the organic carbon (OC) content of the sediment. For the sediments treated by HCl and H_2O, sorption behaviors of Tween-80 fit the linear model very well (R^2: 0.9516–0.9862) at 298 K. The sorption occurred primarily due to partition function of the hydrophobic chains of Tween-80 into the organic carbon of the sediments. Sorption of Tween-80 on H_2O_2-treated sediments followed the Freundlich model (R^2: 0.9565–0.9732), which indicated that the surface function of clay minerals and other inorganic solids in the sediment played a key role. Moreover, the dependence of sorption on salinity and temperature was examined and the thermodynamic parameters were evaluated. It was found that the sorption was favorably influenced by the increasing salinity and decreasing temperature of seawater. The changes of Gibbs free energy ($\Delta G^0 = -15.33 \sim -17.54$ kJ mol^{-1}), enthalpy ($\Delta H^0 = -62.23$ kJ mol^{-1}) and entropy ($\Delta S^0 = -151.68$ J mol^{-1} K^{-1}) of the sorption were also determined. The negative ΔG^0 and ΔH^0 values indicated that the sorption process was spontaneous and exothermic. The negative ΔS^0 value suggested that the degree of freedom decreased during the sorption process.

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1. Introduction

Surfactants are a group of chemicals that contain both polar and nonpolar parts in their structures and are classified as anionic, cationic, nonionic or amphoteric surfactants according to the nature of the hydrophile. Surfactants are widely used in chemical industry, sewage treatment, agriculture and so on. They are applied to a diversity of products including detergents, motor oils, pharmaceuticals and agricultural products (Rosen, 1989). Although they are not regarded as dangerous compounds, most of them will eventually be deposited on the ocean sediments, where they seem to significantly influence the physical, chemical and biological properties of sediments with sorption processes playing a main role (Fytianos et al., 1998). Their accumulation or the accumulation of their biodegradable products comprises an important section of dissolved organic carbon (DOC) in natural water. Meanwhile, they create many health hazards like dermatitis and their eco-toxicological properties lead to destruction of aquatic flora and fauna (Lewis, 1991; Ahel et al., 1993; Field et al., 1995; Khan and Zareen, 2006). Hence their contamination in water systems should not be neglected and has been of major concern in the environmental research regarding their fate, transformation rates (Kile and Chiou, 1989; Liu et al., 1990), toxicological impact (Laha and Luthy, 1991; Guha and Jaffé, 1996) and the influence on the behavior of other hydrophobic organic pollutants (Di Vincenzo and Dentel, 1996; Jones-Hughes and Turner, 2005) in the ocean environment.

In the aquatic environment, sorption to soils and sediments is probably the most influential factor on the transport and fate of organic contaminants (Morel and Gschwend, 1987). With regard to the nonionic surfactants, solid–liquid interaction may be very complicated and for a given sorbate, its sorption behavior can be described by diverse sorption mechanisms, including ion exchange, ion pairing, polarization of π electrons and hydrophobic bonding. The sorption of individual surfactants or surfactant homologues to soils, river sediments, and clay minerals has been studied in some papers (Brownawell and Westall, 1991; Adeel and Luthy, 1995; Cano and Dorn, 1996; Rodríguez-Cruz et al., 2005). But very few information is present in the literature about the sorption behaviors of nonionic surfactants in the seawater–sediment system.

In this paper, the sorption behavior of Tween-80, a representative nonionic surfactant and commercially important one, on marine sediments was systematically investigated. The formula of Tween-80 was shown in Fig. 1. In order to understand the characteristics of transformation of Tween-80 on sediments better and to provide theoretical basis for protecting the ocean environment, we carried out a systematic and comprehensive study on its sorption

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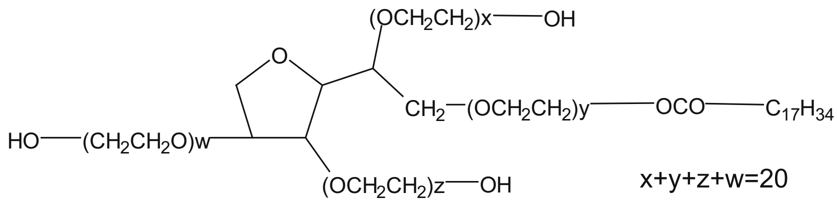

Fig. 1. Formula of Tween-80.

kinetics, sorption isotherms, dependence of the sorption on the salinity and temperature, and finally we estimated the sorption thermodynamic parameters under a wide range of conditions.

2. Experimental

2.1. Sorbents

The sediment samples were collected from three different locations in the Bohai Sea, China. The characteristics of the sediments are shown in Table 1. The organic carbon contents of the samples were determined by the 2400 Series II CHNS/O Analyzer (Perkin–Elmer, USA). The pH values were measured in slurries made at a sediment/water ratio of 1:2.5 (w:w). The mineralogical composition of the sediments was determined by the D/max-rb X-ray diffraction instrument (Rigaku, Japan).

The sediment samples were air-dried in the laboratory and then the shells and other impurities were eliminated. Afterward, they were treated as follows:

A. H_2O-treatment: Sediment samples were only washed with distilled water, air-dried and then grounded. The portion that passed through 40–80 mesh sieves was chosen as experimental sorbents.
B. HCl-treatment: Sediment samples from A were mixed with HCl solution (0.1 M) firstly and then treated according to Zhao et al. (2001). This treatment eliminated the great mass of sediment carbonate (mainly $CaCO_3$ and $MgCO_3$) so that the percentage of OC content of sediment on dry-weight basis increased accordingly.
C. H_2O_2-treatment: The treatment was described in Zhao et al. (2001). This treatment aimed to remove the great mass of the sediment organic carbon. By means of H_2O_2 oxidation, about 90% of the organic carbon in the sediment could be removed (Xu and Li, 2008).

2.2. Seawater

The natural seawater (NSW) collected from the coastal sea of Qingdao was filtered through a 0.45 μm filter membrane and irradiated by strong ultraviolet light to eliminate the effects of natural dissolved organic matter as much as possible (Bester and Hüehnerfuss, 1993; Yang and Zhang, 1997). The salinity and pH values of the natural seawater were 33.4 and 8.07, respectively.

Table 1
Sampling stations of sediments and their characteristics.

Sample	Station	Water depth (m)	Sediment constitutions (%)				OC (%)	pH
			Quartz	Feldspar	Calcite	Clay		
1#	36°N, 120°E	20	54.9	23.5	nd[a]	21.3	0.33	8.87
2#	37°N, 123°E	77	53.3	20	6.3	20.2	0.16	9.38
3#	28°N, 112°E	45	40.5	12.1	7.5	39.1	0.85	9.32

[a] Undetermined.

In order to investigate the effect of the salinity on the sorption behavior of Tween-80, the diluted seawater (DSW) with a final salinity of 16.7 and deionized water (DW) were also chosen as the medium. Furthermore, for the sake of simulating the natural environmental conditions, the pH values of all solutions in the experiments were adjusted to 8.07 by the addition of dilute aqueous solutions of HCl (0.10 M) or NaOH (0.10 M).

2.3. Sorbate and chemicals

Tween-80 ($C_{64}H_{124}O_{26}$) was purchased from Fluka (UK) and used without further purification. Eosin-B was obtained from Shanghai Sangon Biological Engineering Technology & Services Co., Ltd. Other compounds such as HCl and H_2O_2 were AR grade reagents purchased from Shanghai Chemical Reagent Co., China.

The hydrophile–lipophile balance (HLB) number of Tween-80 was 15, indicating it was readily dissolved in the water. The standard solutions were prepared by directly dissolving 0.0500 ± 0.0001 g Tween-80 into 500 mL water (NSW, DSW and DW, respectively) to achieve a final concentration of 100 mg L^{-1}.

2.4. Chemical analysis

The concentration of Tween-80 in aqueous solution was determined according to the modified Al-Kindy protocol (Al-Kindy et al., 2003). This method was based on the theory that fluorescence enhancement will occur if Eosin-B and the surfactant present in the same aqueous solution. Spectrofluorometric analyses were performed on 1.5 mL of Eosin-B solution (20 ppm) added to 1.8 mL of water sample (Andrea et al., 2006). Then the pH value was adjusted to 4.5 by the citrate buffer solutions. Exciting and emission wavelengths were set to 545 and 585 nm, respectively.

2.5. Sorption experiments

For the experiments of sorption kinetics, 0.05 g of the sediment, 25 mL of Tween-80 standard solution and 25 mL of corresponding medium (NSW, DSW and DW) were added to a series of 150 mL Erlenmeyer flasks with a whole suspension volume of 50 mL. The pH of solutions was adjusted to 8.07 ± 0.03 using the HCl or NaOH solutions. In order to keep the pH value constant over the duration of the experiment, the pH value was checked every 4 h and readjusted when it shifted more than 0.5 pH units. Then samples were shaken under a fixed speed (200 rpm) in the dark at 298 ± 0.5 K in a Model SHZ-82 constant-temperature air-bath shaker (Changzhou Guohua Appliance Co., China) at different time intervals as indicated in Fig. 2A and B.

Solid and aqueous phases were separated by a Model SC-3610 centrifuge (USTC Chuangxin Co., Ltd., ZOKIA Branch, China) at a speed of 5000 rpm for 10 min. The supernatant solution was analyzed for surfactant concentration as described above using a Model F-4500 Fluorescence Spectrophotometer (Hitachi Ltd., Japan). All experiments were conducted in duplicate. The adsorbed concentrations of Tween-80 were calculated from the difference between the surfactant concentrations before and after shaking.

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