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Systematic study of the effects of novel halogen-free anionic surface active ionic liquid on interfacial tension of water/model oil system



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ABSTRACT

In this investigation, a novel anionic- halogen-free ionic liquid (IL), 1-butyl-3-methylimidazolium dodecyl sulfate ($[C_4mim][C_{12}SO_4]$), was synthesized and examined for possible application in the chemical enhanced oil recovery (EOR) processes. In this way, the effects of $[C_4mim][C_{12}SO_4]$ and NaCl concentrations, temperature and model oil type (aliphatic and aromatic hydrocarbons) on interfacial tensions (IFTs) between $[C_4mim][C_{12}SO_4]$ solutions and model oils were systematically investigated. It is surprisingly revealed that the novel SAIL $[C_4mim][C_{12}SO_4]$ is highly effective in reducing the IFT of aromatic hydrocarbon/water system, and ultra-low IFT can be achieved in the presence of salinity at higher temperature. Moreover, the interaction energies between $[C_4mim][C_{12}SO_4]$ and different model oil molecules were calculated using molecular mechanics, which could further interpret the reasons of ultra-low IFT.

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1. Introduction

It is well known that there are more than half of crude oil remaining in reservoirs after primary and secondary recovery stages. A large amount of oil is remained unrecovered mainly in the pore structure of reservoirs. Surfactant flooding has been a promising method for mobilizing residual oil from mature reservoirs for many decades. The incremental recovery depends strongly on the capillary number, which is reversely proportional to the interfacial tension (IFT) between the reservoir fluids [1]. Fulcher et al. showed that an increase of capillary number by two or three orders of magnitude can decrease the residual oil saturation of a reservoir by tens of percent [2]. Based on the theory, several groups have systematically studied and evaluated the effects of surfactant flooding in certain reservoir to achieve the high oil recovery efficiency [3–6]. Recently, Liyanage et al. systematically researched that the tristyrylphenol based surfactants have unique structural features that can be easily tailored into enhanced oil recovery (EOR) surfactants [5]. Focus on the EOR of the unconventional reservoirs with more complex conditions, the conventional surfactants cannot satisfy the operating requirements of the harsh environments, therefore it is highly desirable to research the novel high-performance surfactants [7].

Due to the specific chemical and physical properties, the typical organic molten electrolytes Ionic liquids (ILs) have received a good deal

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of attention whose melting point is lower than 100 °C [8,9]. Recently, ILs with the long alkyl chains resembles traditional surfactants and can be referred to as novel surface active ionic liquids (SAILs). The SAILs have been successfully employed in multiple applications because of the specific physical chemical, biological, and electrical properties [10, 11]. To my knowledge, only a few SAILs have been attempted to use in the crude oil industries [12-20], especially for the reduction of IFT in chemical EOR. Hemmateenejad et al. systemically researched the effects of different families (imidazolium and pyridinium) with different chain lengths of SAILs on the IFTs of water/crude oil system [14–16]. Soto et al. have focused on the SAIL-based enhanced oil recovery, mainly for the trihexyltetradecylphosphonium ILs [17-19]. However, all of above reports focus only on the cationic ILs with halogen. Compared to traditional organic solvent, the cationic ILs with halogen are environmentally friendly. But the countra-ion halogen atoms may be insufficiently stable under the hydrolysis or other reactions, releasing corrosive HF or HCl, which can cause massive serious problem [21]. Besides, the absorption loss of the cationic surfactant in formations may significantly restrict their applications in the oil field.

In order to avoid the problems for the typical SAILs consisting of halogen, a mass of halogen-free SAILs with sulfate, nitrite, nitrate, and benzene sulfonate anions have been synthesized [21–24]. For the first time, Wasserscheid et al. synthesized 1-butyl-3-methylimidazolium octyl sulfate ([C_4 mim][C_8SO_4]) [21]. Then most of the reports focused on the phase behavior or the assisted synthesis of nanomaterials [25–30]. Jiao et al. investigated the aggregation behaviors of 1-butyl-3-methyl imidazolium dodecyl sulfate and N-butyl-N-methyl pyrrolidinium dodecyl sulfate in aqueous solution [27]. Due to the well-documented toxicology and biodegradation, the alkyl sulfate-based ILs may be the feasible candidates for the practical applications in various fields [21, 23]. Besides, the absorption loss of the anionic surfactant in formation is obviously in a low level [31]. Recently, Soto et al. synthesized and employed the anionic SAIL tylmethylphosphonium dodecylsulfate to the application of Enhanced Oil Recovery for the first time, decreasing the IFT to 0.08 mN/m [32]. Herein, we introduced the halogen-free anionic SAIL, $[C_4mim][C_{12}SO_4]$ to the new field of chemical EOR, which has been scarcely reported. Then we systematically investigated the effects of several factors on the IFTs between [C₄mim][C₁₂SO₄] solutions and different model oils (aliphatic and aromatic hydrocarbons). It is demonstrated that the SAIL [C₄mim][C₁₂SO₄] more effective to reduce the IFT by the comparison with traditional surfactant and the interaction energy calculation using molecular mechanics.

2. Experimental

2.1. Materials

1-Methylimidazole (>95 wt%) was purchased from ACROS Company. Sodium dodecyl sulfate (SDS > 9 wt%) and sodium dodecyl benzene sulfonate (SDBS > 99 wt%) were purchased from Alfa Aesar. Decane, tetradecane, benzene, hexane and cyclohexane (>98 wt%) were achieved from China Pharmaceutical Group. All other chemicals were of analytical grade and used as received.

2.2. Synthesis of ionic liquid

1-butyl-3-methylimidazolium chloride ($[C_4mim]Cl$) was synthesized according to the literature [23]. 1-butyl-3-methylimidazolum dodecyl sulfate ($[C_4mim][C_{12}SO_4]$) was obtained by ion exchange reaction of SDS and $[C_4mim]Cl$. SDS and $[C_4mim]Cl$ were mixed in dichloromethane for 4 h at room temperature, and the precipitation was filtrated. Then the organic phase was washed using water to the chloride-free in water (verified by the titration of the AgNO₃ aqueous solution), and the white waxy solid $[C_4mim][C_{12}SO_4]$ was obtained. The final products were dried for 48 h in vacuo. The product purity was examined by ¹H NMR spectroscopy with a Bruker Avance 300 spectrometer.

2.3. Measurement of interfacial tension

The interfacial tensions between the model oils and water were measured via spinning drop method on TX-500C spinning drop interfacial tension apparatus (American CNG Company). The aqueous solution was primarily filled to the glass tube. Then a droplet model oil was injected into the centre of the water phase. Finally, the interfacial tension was measured at the fixed rotating velocity (5000 rpm) at the given temperature.

3. Results and discussion

As a typical mixture, the crude oil is composed mainly of aliphatic hydrocarbons, aromatic hydrocarbons, a small number of heteroatomcontaining (N, S, and O) and other uncertain compounds. To eliminate the influence of other impurities, the crude oil was replaced by the pure hydrocarbons. Then we systematically investigated the interfacial activity of the halogen-free SAIL [C₄mim][C₁₂SO₄] between its aqueous solution and aliphatic or aromatic hydrocarbons.

3.1. Effects of $[C_4mim][C_{12}SO_4]$ on the IFTs between hydrocarbons and distilled water

As shown in Fig. 1, the IFTs obviously decreases with the increased concentrations of the $[C_4mim][C_{12}SO_4]$ aqueous solutions (Table S1).

Fig. 1. IFTs between the different hydrocarbons and $[C_4mim][C_{12}SO_4]$ aqueous solutions with different concentrations at 40 °C.

For the aliphatic hydrocarbons (decane and tetradecane), the two curves almost coincides, and both of the IFTs decrease from ~20 mN/ m to ~2.5 mN/m. While the $[C_4mim][C_{12}SO_4]$ is more effective to induce the IFT between its aqueous solution and the aromatic hydrocarbon (benzene) (~20 mN/m to ~0.9 mN/m). To further demonstrate the higher interfacial activity of $[C_4mim][C_{12}SO_4]$, the traditional anionic surfactants with the similar anionic head group (SDS and SDBS) were used in the control experiments when the surfactant concentration was fixed at 3000 mg/L. Interestingly, it is evident that the IFT of $[C_4mim][C_{12}SO_4]$ is much lower than that of SDS or SDBS, especially for the aromatic hydrocarbon (Fig. 2).

It is well known that the addition of sodium halides (e.g. NaCl) can bring dramatic effects to the IFT or surface tension of surfactant aqueous solutions [33]. In our aqueous solutions with no salinity, this difference can be ascribed to the counter-ions due to the similar anionic head group. Due to the preferential adsorption of the surfactant molecules at the interface, the interface with the repulsive charge must delay the adsorption velocity of the subsequent molecules [33]. While the electrostatic interaction between the large surface active group and small counter-ions can promote the movement of the counter ions to the near interface [34]. However, all of the counter-ions are induced to

Fig. 2. IFTs between the different hydrocarbons and the different anionic surfactants with the similar head group aqueous solutions at 40 °C.





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