

The great improvement of the surfactant interfacial activity via the intermolecular interaction with the additional appropriate salt

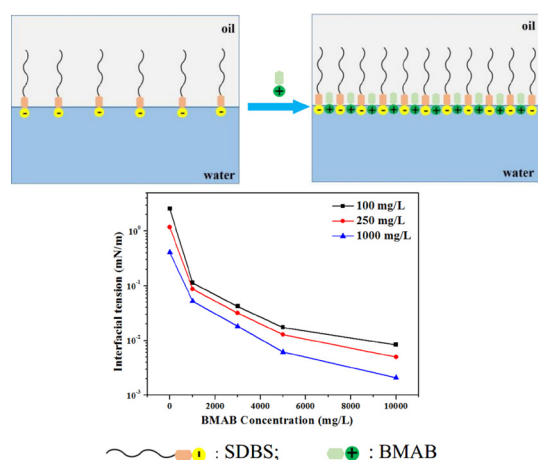


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GRAPHICAL ABSTRACT



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ABSTRACT

The typical organic salt benzyltrimethylammonium bromide (BMAB) was used to greatly enhance the interfacial activity of the traditional anionic surfactant sodium dodecyl benzene sulfonate (SDBS). The additional BMAB caused the obvious reduction of the interfacial tension (IFT) between the SDBS aqueous solution with a low concentration and model oil ($V_{\text{toluene}}:V_{\text{n-decane}} = 1:1$). Based on the special SDBS/BMAB intermolecular interaction, we proposed that the synergistic effect between the electrostatic attraction and π - π stacking interaction largely increased the SDBS interfacial concentration, leading to the dramatic IFT reduction. Three organic salts and two surfactants with similar structures, as well as the typical inorganic salt NaCl, were used in control experiments to confirm our speculation. The interaction energies between SDBS molecules and different organic salts were calculated using molecular mechanics to further interpret the mechanism. The salt and temperature tolerances of the SDBS/BMAB system were systematically investigated. Moreover, the availability of the SDBS/BMAB mixture in brine water/crude oil system was evaluated by the core flooding tests.

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1. Introduction

The displacement efficiency of residual oil is the most significant factor in enhanced oil recovery (EOR). The increase of capillary number, N_c , effectively mobilizes the residual oil trapped in reservoir pore structures, which can be achieved by reducing the water/oil interface tension (IFT) [1,2]. The IFT between crude oil and displacement fluid can be reduced to the ultralow value ($<10^{-2}$ mN/m) by the appropriate surfactant, which remarkably improves the displacement efficiency of oil recovery [3].

To obtain the higher recovery, many groups focused on the investigations of the high-performance surfactants, such as biosurfactants [4–6], gemini surfactants [7,8], zwitterionic surfactants [9–13], surface active ionic liquids [14–16], and mixed surfactants [17,18]. Kamal reviewed the excellent interfacial activities of the gemini surfactants (IFT reduction, wettability alteration, adsorption, and rheology) [7]. Cui's group synthesized the zwitterionic surfactants with two alkyl chains. The large hydrophobic portions caused the enhanced ability to reduce the IFT [9,10]. In short, the novel surfactants with outstanding properties were regarded as the ideal candidates in EOR. However, the complex synthetic methods and correspondingly high cost may seriously restrict their further applications.

The introduction of additive agents is another feasible method to modify the surfactant performances. The cosurfactant alcohols, cyclodextrins, nanoparticles, and polymers were employed in the literatures [19–22]. Recently, the electrostatic attraction between the surfactant head groups and the salts (NaCl, $MgCl_2$, and $CaCl_2$) in brine water was found to increase the interfacial concentrations of surfactant molecules [23–27]. The intermolecular interaction between the NaCl and surfactant molecules were beneficial to the IFT reduction [25]. Further, our group investigated the effects of the intramolecular and intermolecular electrostatic attractions among the surfactant molecules on their arrangements at the water/oil interface [28–31]. Based on the electrostatic attraction and other intermolecular interactions, we successfully fabricated the gemini-like surfactants by combining a single chain surfactant and a bola-type salt at the interface, which exhibited the satisfactory ability to enhance crude oil recovery [29]. Moreover, the counter-ions in surfactant molecules could greatly improve their interfacial activities [28,31]. Enlightened by above results, we speculated the addition of the appropriate salt to the traditional surfactant solution might be a convenient and promising approach to enhance the interfacial properties of the surfactants.

Herein, we added the typical organic salt benzyltrimethylammonium bromide (BMAB) to the traditional anionic surfactant sodium dodecyl benzene sulfonate (SDBS) aqueous solutions. Interestingly, the SDBS/BMAB system could reduce the water/model oil IFT to the ultra-low value at a low SDBS concentration (250 mg/L). Based on the results of control experiments, the intermolecular interactions between SDBS and BMAB, including the electrostatic attraction and π - π stacking interaction, dominated the IFT variations. In addition, the SDBS/BMAB system was further evaluated by the core flooding tests.

2. Experimental

2.1. Chemicals

Sodium dodecyl benzene sulfonate (SDBS, 95%, Fig.S1a), sodium dodecyl sulfate (SDS > 99 wt%, Fig.S1b), dodecyltrimethylammonium bromide (DTAB > 99 wt%, Fig.S1c), benzyltrimethylammonium bromide (BMAB > 99 wt%, Fig.S1d), benzyltriethylammonium bromide (BEAB > 99 wt%, Fig.S1e), tetramethylammonium bromide (TMAB > 99 wt%, Fig.S1f), tetrapropylammonium bromide (TPAB > 99 wt%, Fig.S1g), sodium chloride (> 99 wt%), toluene (> 99 wt%) and n-decane (> 99 wt%) were purchased from Aladdin Chemical Reagent Co. Ltd. The SDBS was recrystallized by methanol, and other chemicals with analytical grade were used as received.

The main compositions of crude oil and the electrolytes in formation brine are given in Table S1 and Table S2, respectively.

2.2. Measurements of interfacial tension

The interfacial tension measurements were conducted on an interfacial tension apparatus (American CNG Company, TX-500C) using the spinning drop method as in our previous reports [28–31]. First, the surfactant solution was filled in the test tube. Then, a small droplet of model oil ($V_{\text{toluene}}:V_{\text{n-decane}} = 1:1$) or crude oil was injected into the water phase. The volume ratio of water/oil was approximately 200. The rotating velocity in the measurements was set at 8000 rpm at the given temperature.

2.3. Core flooding tests

In the typical flooding test, the artificial core was 60 cm in length and 5.0 cm in diameter, which was from a sandstone reservoir. The tests were performed at the oil reservoir temperature of 65 °C. First, the core was saturated with the brine, followed by the injected crude oil to almost no generated brine (brine cut < 1%). Next, the core was flooded with the brine until no more oil was produced (oil cut < 1%). Then, a 0.5 PV slug of the brine solution containing the chemical compositions was injected. Finally, an extended brine slug was injected until the oil cut was negligible. The injected rate was given at 2.0 mL/min.

3. Results and discussion

3.1. Effects of additional BMAB molecules on the IFT between model oil and SDBS aqueous solutions

Fig. 1 presents the decreased tendencies of three IFT curves with the increased BMAB concentrations. The slight additional BMAB molecules (1000 mg/L) obviously decrease the IFT values between model oil and SDBS aqueous solutions by one order of magnitude, and the ultra-low IFT values ($<10^{-2}$ mN/m) are obtained at high BMAB concentrations (5000 mg/L ~ 10,000 mg/L). To confirm the effects of the typical organic salt BMAB, the traditional inorganic salt NaCl was used to replace BMAB in the control experiment. As shown in Fig. 2, the additional NaCl merely causes the moderate IFT reduction. At the same salt concentration, the IFT value for SDBS/BMAB system is about two orders of magnitude lower than that for SDBS/NaCl system. Due to its greater molar mass, the BMAB molecular number is much less than that of NaCl.

For the ionic surfactant, the electrostatic repulsion among the charged head groups of the surfactants prevents their close-packed

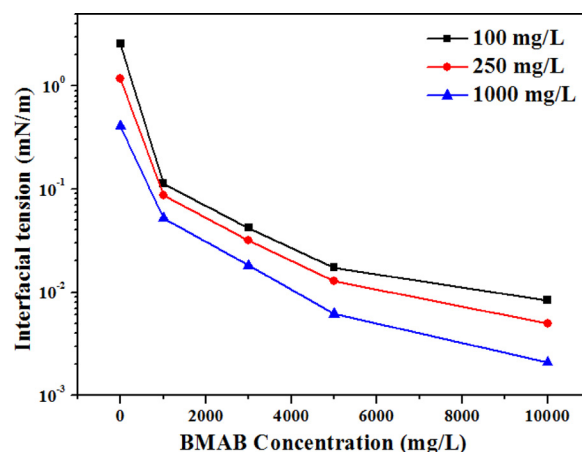


Fig. 1. Effects of additional BMAB molecules on the IFT between model oil and SDBS aqueous solutions (100 mg/L, 250 mg/L, and 1000 mg/L) at 30 °C.

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