



Effect of polymer on dynamic interfacial tensions of sulfobetaine solutions

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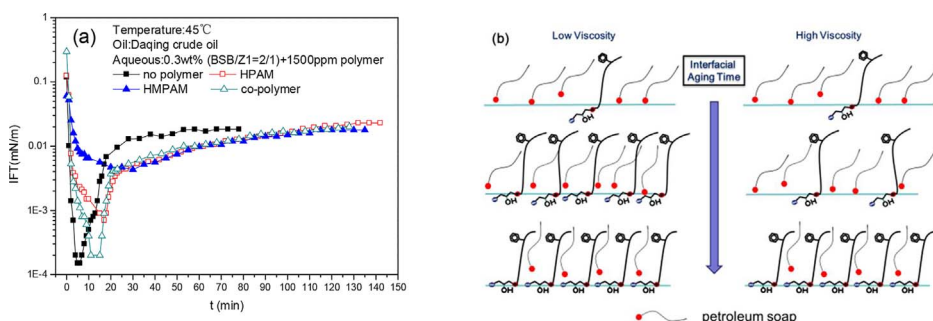
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GRAPHICAL ABSTRACT

Polymer slows the diffusion-adsorption of betaine at the interface and results in the disappearance of compact mixed adsorption film with the aging time.



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ABSTRACT

The effect of different types of polymers, partly hydrolyzed polyacrylamide (HPAM), hydrophobically modified polyacrylamide (HMPAM) and co-polymer, on the dynamic interfacial tensions (IFTs) of benzyl substituted alkyl sulfobetaine (BSB) against n-alkanes, kerosene and crude oil have been investigated by a spinning drop interfacial tensiometer. The influence of polymer concentration on IFTs was expounded. The experimental results show that betaine has high interfacial activity and the addition of polymer has great influence on both dynamic IFT behaviors and equilibrium values between surfactant solution and n-alkanes mainly by modifying the interfacial molecular arrangement on the interface. However, the addition of polymer affects the dynamic IFTs of surfactant solution against kerosene and crude oil, but has little effect on the equilibrium IFT values. Active substances existing in kerosene and the in situ produced petroleum acidic soaps play a synergistic effect with betaine to reduce the IFTs. The increase of viscosity of aqueous phase caused by polymer influence the adsorption of the surfactant at the interface, slowing down the decrease of IFTs and changing the minimum transient IFT values.

1. Introduction

The technology of enhanced oil recovery (EOR) has become more and more important because approximately two-thirds of the oil has been left in an average oil reservoir after the primary and secondary production modes, which is believed to be in the form of ganglia and

trapped in the pore structure of the rock by capillary forces. The capillary number, N_c , determines the microscopic displacement efficiency of oil trapped in the reservoir pore structure and is important to the oil recovery by a chemical flooding process. Larger the capillary number N_c , higher the oil recovery efficiency. [1] N_c is directly proportional to the viscosity of the displacement fluid, and varies inversely with the

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interfacial tension (IFT) between the displacement fluid and crude oil. Increasing the viscosity of the displacement fluid and reducing the oil/water interfacial tension are the most effective ways to improve the Nc value. The IFT between oil and aqueous solution can be reduced dramatically by the employment of an appropriate surfactant system. The addition of the polymer greatly increases the viscosity of aqueous phase. Therefore, surfactants and polymers have been widely used in EOR. [2–5]

Chemical flooding includes surfactant flooding, polymer flooding, alkaline-polymer flooding, surfactant–polymer (SP) flooding, and alkaline-surfactant–polymer (ASP) flooding. [6–8] With the advantages of low operation cost and wide range of use in oil fields, SP has received more attention. [9,10] Compared with conventional anionic, cationic and nonionic surfactants, zwitterionic surfactants have special molecular structure of both cationic and anionic centers attached to the same molecule. Generally, zwitterionic surfactants can exist as zwitterions over a wide pH range and show many unique properties. High foam stability, temperature resistance and salt tolerance make it more suitable for harsh conditions. Low toxicity and biodegradation meet the requirements of environmental protection. [11–13] Betaines are an important kind of zwitterionic surfactants and have wide applications. Recently many studies have focus on the synthesis and interfacial properties of betaine applied to EOR. [12,14]

Partially hydrolyzed polyacrylamide (HPAM) is widely used as thickener or modifier in the areas of tertiary oil recovery, drilling fluids and many other processes. [15] With the development of oil fields, the temperature and salinity in the reservoirs are getting higher and higher, which put forward much higher requirements for polymer. Another important EOR polymer is hydrophobically modified polyacrylamide (HMPAM), modified polyacrylamides with relatively low amounts of hydrophobic co-monomers. HMPAM is also used as aqueous viscosity modifier but has become the subject of extensively research with their interesting solution behavior. [16]

The influences of polymers on the interfacial tension have been reported widely. Recently, Ma et al. investigated the anionic surfactant/polymer/model oil system and found that the addition of polymers mostly resulted in an increase of IFT because the interfacial molecular arrangement was modified owing to the interaction between polymer and surfactants. [17] Zhu et al. found the similar influence of polymers on anionic–nonionic surfactant/n-decane system and polymers show little effect on the IFTs of anionic–nonionic surfactant system with longer EO chain because of the steric hindrance. [18] Li et al. studied the dynamic interfacial tensions of surfactant/polymer/organic alkali systems against a homologous series of alkanes and found that HPAM will obviously enhance the water solubility of the surfactant/organic alkali solutions and reduced the IFT values against hydrocarbons with lower alkyl carbon number. [19]

Unlike other types of surfactants, betaine has a unique way of adsorption, according to our previous study [20–23]. However, the influence of the polymer on the IFT of zwitterionic surfactants is rarely reported. So it is very interesting and necessary to study the effect of the polymer on betaine. In this paper, the effect of three types of polymers (HPAM, HMPAM and co-polymer) on the IFT of benzyl substituted alkyl sulfobetaine solutions against n-alkanes, kerosene and Daqing crude oil has been investigated. It may be useful for us to understand the mechanisms responsible for the IFT behaviors of zwitterionic surfactant solutions and has great important guidance for the design of EOR formulation with polymer.

2. Experimental

2.1. Materials

In this study, benzyl substituted alkyl sulfobetaine (BSB) with the mass fraction of 33% was synthesized in our laboratory. The structure is plotted in Fig. 1. The basic surface-active parameters of BSB are listed

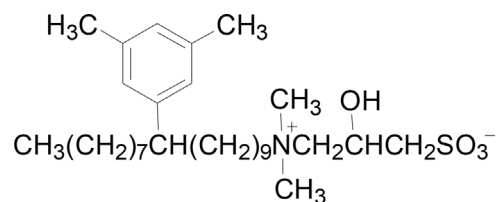


Fig. 1. Structure of Benzyl Substituted Alkyl Sulfobetaine.

elsewhere. [22] Z1 is a polyether nonionic surfactant used for the sake of dissolution of BSB. The alkanes (> 99 mol%) have chain lengths from C6 to C14. Kerosene was further purified by glass chromatography column filled with the silica gel layer until the IFT value is about 40 mN/m against pure water. Partly hydrolyzed polyacrylamide (HPAM) with 20.0% degree of hydrolysis and viscosity average molecular weight of about 1.4×10^7 , hydrophobically modified polyacrylamide (HMPAM) with viscosity average molecular weight of about 1.0×10^7 and co-polymer (HPAM with viscosity average molecular weight of about 4.0×10^6) were obtained from Daqing Oilfield, China. The crude oil was from Daqing oil field with a density of 0.850 g/cm³ at 45 °C. All of the aqueous solutions used in this study were prepared with formation brine. The compositions of the formation brine were listed in Table 1. The pH values of formation brine, surfactant solutions and surfactant-polymer solutions were listed in Table 2. The viscosity of BSB and Z1 solution is 1.05 mPa s, and the viscosity of surfactant-polymer solutions are shown in Table 3.

2.2. Apparatus and methods

The dynamic interfacial tensions were measured by a Texas-500C spinning drop interfacial tensiometer (CNG USA CO.). The standard spinning-drop tensiometer had been modified by the addition of video equipment and an interface to a personal computer. The computer had been fitted with a special video board and a menu-driven image enhancement and analysis program. The video board can “capture” a droplet image for immediate analysis. Analysis usually consists of measurement of drop length and drop width.

The surfactant solution as an outer phase was injected into the glass tube, and about 2 μL of oil as an inner phase was put into the middle of the tube. In all cases, the measurements of the interfacial tension are at a rotating velocity of 5000 rpm. The volumetric ratio of water to oil in the spinning drop tensiometer is about 200. Samples were assumed to be equilibrated when measured values of IFT remained unchanged for half an hour. All experiments were performed at 45.0 ± 0.5 °C.

3. Results and discussion

3.1. Dynamic IFTs between BSB solutions and n-alkanes

As known, there is great dissimilarity in the natures of oil and water. For displacement of the oil in the pores and capillaries of petroleum reservoir rock, the IFT value of 10^{-3} mN/m order of magnitude is generally required. The ultralow IFT can be reached by almost complete replacement of solvent molecules at the interface by surfactant molecules.

Mixed surfactant solution (BSB/Z1, mass ratio of BSB and Z1 is 2/1, for the sake of dissolution of BSB) was selected as model surfactant. The IFTs of BSB solution and Z1 solution against n-decane were also studied to understand the difference of interface activity between BSB, Z1 and their mixture. Fig. 2 illustrates the effect of surfactant concentration from 0.001 to 0.5 mass% on the dynamic IFTs against n-decane. The dynamic IFTs of BSB and Z1 have the same trends as that of their mixture, so they aren't listed for the sake of brevity. It can be seen from Fig. 2 that the IFTs continuously decrease and then reach a plateau along with time, and the curves show typically a L-shaped. [24] The

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