



Effect of electrolyte on interfacial dilational properties of chemical flooding systems by relaxation measurements

Yang-Wen Zhu^{a,b}, Lei Zhang^b, Xin-Wang Song^b, Lan Luo^c, Lu Zhang^{c,*}, Sui Zhao^c, Jia-Yong Yu^c

^a State Key Lab. of Oil and Gas Reservoir Geology and Exploitation, Southwest Petroleum University, Chengdu City, Sichuan 610500, China

^b Geological Scientific Research Institute of Shengli Oilfield Co. Ltd., SINOPEC, Dongying, Shandong 257015, China

^c Technical Institute of Physics and Chemistry, Chinese Academy of Sciences, Beijing 100190, China

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ABSTRACT

In the present work, the influences of different types of electrolytes on interfacial dilational properties of anionic surfactant sodium 4,5-diheptyl-2-propylbenzene sulfonate 377 and asymmetrical anionic Gemini surfactant $C_{12}COONa$ - p - C_6SO_3Na in the absence or presence of 1500 ppm partly hydrolyzed polyacrylamide were studied at decane–water interface, respectively, by means of interfacial tension relaxation measurements. The decay curves of interfacial tension were fitted by the summation of a number of exponential functions. The dilational elasticity (ϵ_r) and dilational viscosity component (ϵ_i) were calculated by Fourier transform and displayed as Cole–Cole plots (plotting ϵ_i or ϵ_i/ϵ_0 as a function of ϵ_r or ϵ_r/ϵ_0 respectively). The experimental results show that only single reorientation process dominates the interfacial properties in the presence of any electrolyte for 1×10^{-6} mol L⁻¹ 377 solution, resulting from the more compacted film by electrostatic screening. On the other hand, the contribution of reorientation process at higher frequency decrease after the addition of NaCl and there exists no pure reorientation process in the presence of $CaCl_2$ or $MgCl_2$ for Gemini surfactant because of the slight increase of interfacial concentration due to larger molecular size and strong steric hindrance between alkyl chains. The addition of polymer can significantly modify the dilational properties of adsorbed surfactant layer due to the formation of mixed adsorption film through hydrophobic interaction between polymer chain and alkyl chain of surfactant molecule. The normalized Cole–Cole plots of surfactant–polymer films with different types of electrolytes show the similar characteristic in general and no pure reorientation process can be observed in any case.

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1. Introduction

It is well known that crude oil plays an important role in providing the energy supply of the world among various sources of energy. At the end of water flooding, almost 65–70% of OOIP is left in the reservoirs. In order to recovery additional oil by a chemical flooding process, the capillary number, which determines the microscopic displacement efficiency of oil, should be increased by 3–4 orders of magnitude through reducing the interfacial tension (IFT) of oil ganglia from its value of 20–30 mN/m to 10⁻³ mN/m [1]. It has long been recognized that the IFT between crude oil and chemical flooding solutions can be reduced to lower than 10⁻² mN/m by using an appropriate surfactant system. Moreover, surfactant–polymer flooding can expand volumetric sweep, but also improve the wash oil efficiency, which is considered one of the most commercial prospects in Enhanced Oil Recovery (EOR) [2].

* Corresponding author. Tel.: +86 10 82543587; fax: +86 10 62554670.

E-mail address: luyiqiao@hotmail.com (L. Zhang).

The physico-chemical environment of petroleum reservoirs can influence the behavior of oil recovery processes and the main elements of the reservoir environment, such as the in-place oil and water, the mineralogy, the geology/lithology and the temperature, have been discussed in detail early [3]. Among these elements, the interstitial water is the major factor controlled the behavior of chemical flooding solutions. Certain chemical constituents in the interstitial water can strongly influence the injection sequence of the fluids, the longevity or attenuation of the flooding slugs, the efficiency of the slug in mobilizing oil, the mobility of the chemical flooding solutions. The concentration of both mono- and divalent metal ions can affect the interfacial tension, viscosity and phase stability of the solutions. For an example, laboratory studies have shown that the required large reduction in interfacial tension can only be achieved in a very narrow range of slat concentration due to the partitioning equilibrium of surfactant molecules between the oil phase and the aqueous phase is sensitive to salinity [4].

The interfacial dilational rheological properties are believed to be very important for controlling the stability of foams and emulsions, consequently dominate the formation of oil bank during

flooding process and de-emulsification process [5–8]. Some rheological studies of water/crude oil or water/model oil interfaces have been made to clarify the mechanisms involved as related to EOR [9–21]. The rheological properties of these interfaces have been found to be strongly dependent on the nature of solvent used for dilution, the oil concentration, the asphaltene and resin concentrations, the resin to asphaltene ratio, and so on. However, practically little attention has been drawn to the effect of electrolyte on the viscoelastic properties of adsorption layers of the flooding systems.

Interfacial pressure relaxation method is a useful way to study interfacial dilational properties of adsorption films, especially for systems controlled by slow relaxation processes. This method has been successfully employed in interfacial films formed by proteins and polymers [22–24]. In the present work, aiming at the interfacial phenomena occurring in EOR process, the influences of mono- and divalent metal ions on the interfacial dilational properties of different types of surfactants (anionic surfactant sodium 4,5-diheptyl-2-propylbenzene sulfonate 377 and asymmetrical anionic Gemini surfactant $C_{12}COONa$ -p- C_9SO_3Na) and polymer (partly hydrolyzed polyacrylamide HPAM) systems were studied, respectively, by means of interfacial tension relaxation measurements.

2. Theory

The Gibbs interfacial dilational modulus is defined by the surface tension increase after a small increase in area of a surface element:

$$\varepsilon = \frac{d\gamma}{d \ln A} \quad (1)$$

It gives a measure of the interfacial resistance to changes in area. Where ε is the dilational modulus, γ is the interfacial tension and A is the interfacial area. The dilational modulus can also be defined as a complex function which can be written as: [25,26].

$$\varepsilon = \varepsilon_d + i\omega\eta_d \quad (2)$$

where ε_d is the dilational elasticity or storage modulus and ε_η the dilational viscosity component or loss modulus that represents a combination of internal relaxation processes and relaxation due to transport of matter between the surface and the bulk.

Interfacial tension relaxation experiments are a reliable way to obtain surface dilational parameters, which uses small but fast axisymmetric trough or drop area expansion or compression to slightly disturb the monolayer equilibrium (In general, the interface was assumed to be equilibrated when the surface tension did not change with time). This causes an interfacial tension jump and then the interfacial tension will decay to the equilibrium again.

Phase angle θ , describing the phase difference between dynamic interfacial tension variation and interfacial area variation, is calculated according to.

$$\tan \theta = \frac{\varepsilon_\eta}{\varepsilon_d} \quad (3)$$

For an instantaneous area change rising from $\Delta A(t) = 0$ for $t \leq 0$ to $\Delta A(t) = \Delta A$ for $t > 0$, the values of ε are obtained as a function of the frequency by Fourier transformation (FT) of the interfacial tension decay obtained from the experiment by the following relationship [27–29]:

$$\varepsilon(\omega) = \frac{FT\Delta\gamma(t)}{FT(\Delta A/A)(t)} = \frac{\int_0^\infty \Delta\gamma(t) \exp(-i\omega t) dt}{\int_0^\infty [\Delta A(t)/A] \exp(-i\omega t) dt} \quad (4)$$

where ω is the angular frequency.

For a real system a number of relaxation processes may occur and the decay curve would be expressed by the summation of a number of exponential functions: [22,23].

$$\Delta\gamma = \sum_{i=1}^n \Delta\gamma_i \exp(-\tau_i t) \quad (5)$$

where τ_i is the characteristic frequency of the i th process; $\Delta\gamma_i$ is the fractional contribution which that relaxation process makes to restore the equilibrium; n is the total number of the relaxation processes.

And the dilational elasticity ε_d and the interfacial dilational viscosity component $\omega\eta_d$ are given by

$$\varepsilon_r(\omega) = \varepsilon_d(\omega) = \frac{\omega}{\Delta A/A} \int_0^\infty \Delta\gamma(t) \sin(\omega t) dt \quad (6)$$

$$\varepsilon_i(\omega) = \omega\eta_d(\omega) = \frac{\omega}{\Delta A/A} \int_0^\infty \Delta\gamma(t) \cos(\omega t) dt \quad (7)$$

Other parameters such as the tangent of phase angle, dilational modulus etc. can all be obtained from these two parameters.

3. Experimental section

3.1. Materials

The surfactant sodium 4,5-diheptyl-2-propylbenzene sulfonate (377) and the asymmetrical anionic gemini surfactant $C_{12}COONa$ -p- C_9SO_3Na were synthesized by ourselves [30,31], as showed in Scheme 1. The purity of the compounds was checked by elemental analysis and 1H NMR spectroscopy. The critical micelle concentrations (CMC), which are taken as the concentration at the point of intersection of the two linear portions of the γ -log C plot, are 1.27×10^{-4} mol/L and 1.10×10^{-4} mol/L for 377 and $C_{12}COONa$ -p- C_9SO_3Na respectively. The polymer, partly hydrolyzed polyacrylamide Mo-4000, was supplied by Mitsubishi Corporation (Japan) with hydrolysis of 25.0% and active content of 90.0%. Its viscosity average molecular weight is about 22×10^6 . Decane, A.R., was obtained from Xingjin Chemical Reagent Ltd., Tianjin, China and used as oil phase without further treatment. Sodium chloride, Calcium chloride and Magnesium chloride were all A.R. and purchased from Beijing Fine Chemical Industry Co., Ltd., China. Water used in the experiments was distilled twice from potassium permanganate solution.

3.2. Methods

The interfacial dilational viscoelasticity meter JMP2000A (Powereach Ltd., Shanghai, China) which had been described elsewhere [32], was employed. The apparatus is designed to monitor the interfacial tension and area changes occurring on the instantaneous expansion of an interfacial film. After perturbation there is a rearrangement of surfactant molecules to reestablish the equilibrium state, i.e., an interfacial relaxation occurs. The working principle is similar to that of Lucassen and Giles. A Langmuir trough consists of a pair of symmetrically oscillating PTFE barriers for changing the interfacial area. The dynamic interfacial tension was measured by the Wilhelmy plate method, using a PTFE plate suspended in the middle of the trough area from a sensitive force transducer. The water phase (90 mL) and oil phase (50 mL) was poured into the trough successively and carefully. The Wilhelmy plate should be completely submerged under the surface of the oil phase. After 6 h of pre-equilibrium of the oil–water system, the interfacial tension relaxation measurements were carried out (the film was expanded about 10% in area by a sudden expand in 2 s). In the interfacial tension range of the studied systems, the

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