Journal of Petroleum Science and Engineering 168 (2018) 283–300

Contents lists available at ScienceDirect

Journal of Petroleum Science and Engineering

journal homepage: www.elsevier.com/locate/petrol

Characterization of alkali-surfactant-polymer slugs using synthesized gemini surfactant for potential application in enhanced oil recovery

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ARTICLE INFO

Keywords:
Gemini surfactant
Interfacial tension
Thermal stability
Viscosity
Viscoelasticity
ASP flooding

ABSTRACT

The present study deals with the formulation and characterization of alkali-surfactant-polymer slugs using gemini surfactant (GS) for effective application in enhanced oil recovery (EOR) processes. The hydrolytic stabilities of the synthesized cationic GSs were measured in both acidic and basic media. Surface tensiometry was employed to determine the CMC values of surfactant at different temperatures. An organic alkali, monoethanolamine (MEA) showed the most favourable interfacial properties at the oil-aqueous interfaces among the various experimented alkalis. Though the addition of PHPA polymer to GS/MEA solutions increase the IFT value, the viscosities of the slugs improve drastically, which is desirable to improve the sweep efficiency. The solutions also showed low turbidity values and high thermal stability over a significant time period. The oil-surfactant-aqueous systems showed favourable emulsification properties, showing the ability to effectively displace trapped oil. Viscosity increases with PHPA concentration due to mixed micelle associations among the surfactant, alkali and polymer molecules. At low shear rates, the GS/MEA/PHPA solutions exhibited shear thinning properties. Oscillatory studies show the presence of both viscous and elastic properties, which have been well described using the Maxwell mechanical model. Formulation consisting of 0.02% 14-6-14 GS, 0.80% MEA and 0.20% PHPA showed the most favourable tertiary recovery after conventional secondary water-flooding.

1. Introduction

Primary and secondary recovery techniques account for the recovery of about one-third of the original oil in place (OOIP) in existing reservoirs. The residual oil remains trapped within the rock pores and can be extracted by tertiary or enhanced oil recovery (EOR) techniques (Druetta et al., 2017; Leray et al., 2016). One of these methods incorporate the use of chemicals with varying compositions to achieve the objective of maximizing oil recovery (Liu et al., 2017; Yusuf et al., 2016). Most injection fluids used as slugs for chemical flooding techniques generally consist of a mixture of surfactants, polymers and alkalis (Hosseini-Nasab et al., 2016; Olajire, 2014; Shen et al., 2009). They are generally employed in significant quantities and, hence, in most cases, expensive. Currently, the oil industry has focused on addressing these problems by pursuing cost-effective research in the field of EOR (Bai et al., 2017; Negin et al., 2017). This has, in turn, generated rapid interest among researchers to develop and optimize surfactant-alkali-polymer formulations that recover more oil with less cost. A relatively new class of surface active agents known as gemini (or dimeric) surfactants has the potential to make significant strides in this field. A gemini surfactant (GS) has two hydrophilic head groups and two alkyl hydrophobic tails, as opposed to a conventional monomeric surfactant with a single polar head and a single tail (Sheng, 2015; Zhao and Wang, 2017). Their critical micelle concentration (CMC) values are extremely low due to the ability of GS molecules to self-aggregate at low concentrations (Sharma et al., 2017; Vongsetskul et al., 2009). Greater the hydrophobicity, greater is the ability of the surfactant to attract the trapped oil in the reservoir. GSs may be highly efficacious in this regard due to the presence of two hydrophobic (alkyl) entities in a single gemini molecule. The selection of proper GS for formulation design is based on a number of factors such as surfactant type, gemini spacer length, relative hydrophobicities and micellization tendencies in the aqueous phase (Hussain and Kamal, 2017; Zhang et al., 2014). Most studies pertaining to the application of gemini surfactants in EOR is focussed on the evaluation of interfacial tension, stability, rheology as well as flooding tests of surfactant slugs only (Kamal et al., 2017; Negin et al., 2017; Pal et al., 2017a,b; Yuan et al., 2017). A favourable rheological characteristics have also been observed, which improve the mobility of displaced oil in reservoirs (Kamal et al., 2017; Negin et al., 2017). Their unique and versatile physicochemical traits contribute towards their potentiality in oil recovery operations.

Alkali-surfactant-polymer systems have been highly successful in

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https://doi.org/10.1016/j.petrol.2018.05.026
Received 11 January 2018; Received in revised form 28 March 2018; Accepted 7 May 2018
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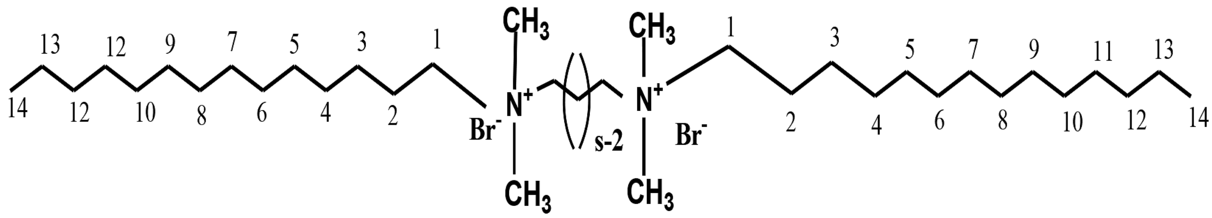

Fig. 1. Molecular structure of 14-s-14 quaternary GS molecule.

chemical EOR processes over the last few decades (Carrero et al., 2007; Ding et al., 2016; Li et al., 2015). However, the effect of alkali and polymer addition portraying the effect of alkali-surfactant-polymer (ASP) formulations on oil recovery is not very extensive (Gao, 2012; Weerasooriya et al., 2017). With development of existing oil fields, the mineralized water percentages in reservoir formations are improved, which creates a need for complex mixtures with varying compositions of surfactant, polymer and alkali that may prove to be highly beneficial. Though sand adsorption issue is common in case of cationic surfactant application in EOR, the synthesized surfactants possess very low IFT values in comparison to commercial surfactants. At high surfactant concentrations, adsorption increases very slowly, that does not cause significant damages in oil displacement ability of injected slug (Zana and Xia, 2004; Zhou et al., 2017). Therefore, if a high enough surfactant concentration is employed in EOR, their rock adsorption property can be effectively controlled. Alkali reacts with the crude oil acids to form in-situ soaps and consequently, reduce the IFT at the oil-aqueous interfaces (Dehghan et al., 2015; Yuan et al., 2015). Though polymer addition reduces the IFT value of the solution, rheological properties are remarkably improved. Viscosity improvement aids in increasing the ability of the displacing (injected) phase to effectively mobilize the oleic phase trapped in the reservoir (Pal et al., 2016; Radnia et al., 2017). In this paper, an attempt has been made to discuss as well as formulate effective ASP solutions that can be applied as effective injection slugs in chemical EOR processes. Ammonium bromide–based gemini surfactants show improved interfacial and oil solubilizing characteristics in comparison to non-ionic gemini surfactants (Goloub et al., 1996). Non-ionic gemini surfactants synthesized from sugars, carbohydrates, etc. have been reported to achieve reduced adsorption onto the sand surface (Fan et al., 2013; Guo et al., 2012). However, their solubilities as well as interfacial tension are not very favourable and, hence, are used in mixed systems (Ahmadi and Shadizadeh, 2012). At the injection stage, the solution slug must exhibit pseudoplastic behavior in order to achieve better injectivity (Mohajeri et al., 2015). Once it is introduced far into a reservoir, it remains exposed continuously to the dynamic high shear rate conditions (Li et al., 2016). During this period, the displacing fluid viscosity must be high enough to effectively control the mobility of the oleic phase. This improves the oil displacement efficiency of the injected phase under reservoir conditions. Viscoelasticity renders improved flow of trapped crude oil in heterogeneous reservoirs, where permeability variations are significant (Santvoort and Golombok, 2016; Sharafi et al., 2018). The ability of a fluid to show both viscous and elastic characteristics, when undergoing deformation, aids in the sweeping of oil at the pore ends as well as at the pore throats. Gemini surfactants also exhibit long-term emulsion stabilities, which effectively improves mobility control in reservoirs with variable permeability zones and improve oil extraction ability (Liu et al., 2012; Yuan et al., 2017). Thus, it is pertinent to maintain an intricate balance among various physicochemical properties of slug compositions in order to design an optimized alkali-surfactant-polymer (ASP) formulation with functional application and cost-efficiency in EOR operations.

In this paper, the physicochemical properties of GS-alkali-polymer systems were investigated in order to develop a perfect formulation for application in oil recovery processes in mature reservoirs. Initially, the hydrolytic stabilities of 14-s-14 GSs (s = number of aliphatic carbon atoms in spacer of gemini molecule) were studied in determine its degradability characteristics in acidic and basic media. The CMC values of different surfactants were measured by surface tension measurements. Interfacial behavior between crude oil and aqueous solution was studied by spinning drop method in order to understand the relative effects of surfactant, alkali and polymer on IFT. Thermal stabilities were analyzed for formulations with varying compositions to check their functionality under high temperature conditions. Emulsification abilities of surfactant/alkali systems were measured to investigate the relative efficacies of different GSs in oil recovery processes. The formulations were also characterized by rheological experiments under different conditions. Viscosity as well as viscoelastic moduli were studies as a function of polymer concentration and temperature in ASP solutions. Flooding tests were finally conducted to determine the oil recovery percentages achieved by chemical slugs, after conventional water-flooding.

2. Materials and methods

2.1. Materials

The crude oil employed in this paper was obtained from Ahmedabad oil field. The oil has a total acid number (TAN) of 0.044 mg KOH/g, kinematic viscosity of 6.147×10^{-5} m^2/s and gravity of 23.55° API at 303 K. Alkalis, i.e. monoethanolamine (MEA), diethanolamine (DEA), triethanolamine (TEA) and sodium hydroxide (NaOH) were purchased from Rankem Chemicals. Double distilled water (DDW) was extracted from distillation apparatus. The surfactants employed in this study possess a dimeric structure and are essentially gemini surfactants (GSs) as shown in Fig. 1. Therefore, their molecular structures consist of two polar head groups and two alkyl hydrophobic tails. It is noteworthy mentioning that the GSs were produced in significant quantities in the laboratory to conduct various experiments in this study. The synthesis and characterization results of the four GSs have been discussed in our previous papers (Pal et al., 2017a,b).

The ^1H NMR spectra of the synthesized GSs, calculated from Bruker AVANCE III 500 MHz (AV 500) multi nuclei solution NMR Spectrometer using heavy water (D_2O) lock solvent, are shown in Fig. 2. The different peaks obtained for each GS specimen are described in Table 1. Analysis of ^1H NMR data confirm that the analyzed compounds have a dimeric structure, comprised of two alkyl tails (with fourteen straight-chain carbon atoms) attached to quaternary ammonium groups on either side, connected with each another with a polymethylene spacer group. Using combustion technique, the specimens were evaluated using a FLASH EA 1112 series CHNS(O) analyzer (Thermo-Finnigan, Italy). As per the principle of Dumas method, combustion products were separated by column chromatography and then detected by thermal conductivity meter (TCD). The elemental compositions of the synthesized GSs in terms of percentages of carbon, hydrogen and nitrogen are shown in Table 2. Quantitative elemental studies using CHN (Carbon-Hydrogen-Nitrogen) method prove that 14-3-14, 14-4-14, 14-5-14 and 14-6-14 GSs synthesized in the laboratory possess purities of 75.94%, 76.10%, 76.41% and 78.30% respectively.

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