Contents lists available at ScienceDirect

Fuel

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

Full Length Article

Flow regimes during surfactant flooding: The influence of phase behaviour

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ARTICLEINFO

Keywords: Microfluidics Displacement mechanisms Enhanced oil recovery Surfactant flooding Phase behaviour Microemulsion

ABSTRACT

Surfactant flooding is one of the most widely adopted enhanced oil recovery strategies whereby microemulsion is formed *in situ* during immiscible displacement. It is expected that local equilibrium is achieved during surfactant flooding and that resulting microemulsion phase properties directly influence the flow physics and thus, oil recovery. We consider phase behaviour of a surfactant-oil-water system that can form either Winsor type II-, III or II+ microemulsion. Water, polymer and Winsor type surfactant solutions are injected into oil saturated polydimethylsiloxane microfluidic chips and imaged under continuous flow at various Capillary numbers and Viscosity ratios. Images are analysed for displacement patterns and oil recovery. Flow regimes are explained by considering the equilibrium interfacial tension (IFT) and viscosity of the formed microemulsion phase. Displacement of oil by injected fluids was dominated by capillary fingering at low flow rates. At higher flow rates, type III microemulsion develops viscous fingering while type II – develops stable displacement due to the microemulsion phases being of high and low viscosity, respectively. This report highlights that the difference between stable/unstable displacement during surfactant flooding is influenced by surfactant-oil-water phase behaviour.

1. Introduction

During oil recovery from a typical hydrocarbon reservoir less than half of the original oil in place is recovered due to capillary trapping and reservoir heterogeneity [1]. Subsequently, enhanced oil recovery (EOR) processes are required to recover the additional oil [2–4]. Surfactant flooding is an EOR approach that works by lowering the interfacial tension (IFT) between brine and oil and thus, targeting capillary trapped oil [1,5]. With surfactant flooding, microemulsions (or emulsions) are formed *in situ* and IFT values can be ultralow ($< 10^{-2}$ mN/ m) [6,7]. However, in most cases, there is still an interface between phases and thus, displacement remains immiscible [1,8].

To better understand ultra-low interfacial tension displacement, we can start by reviewing what is known about standard immiscible displacement. One cause for inefficient oil recovery is due to instabilities that can occur during fluid/fluid displacement [9,10]. These instabilities arise from the competition between viscous and capillary forces and have been studied extensively with microfluidic devices and numerical simulations [10–14]. Immiscible two-phase displacement is often characterised by two dimensionless parameters: Capillary number (*Ca*) and Viscosity ratio (v) [10,15]. *Ca* shows the effect of viscous to interfacial forces, which is represented as

$$Ca = \frac{\mu u}{\sigma} \tag{1}$$

where μ is displacing fluid viscosity (Pa·s), u is average pore velocity (m/s), and σ is the interfacial tension (N/m) between the displaced and displacing fluids. Another important parameter is Viscosity ratio (v), which is defined as the ratio of displacing fluid viscosity to displaced fluid viscosity. For v > 1, the displacement is considered favourable and instabilities such as viscous fingering do not occur [10,16]. In addition, relative permeability is another factor that influences stability during immiscible displacement in porous media. Several attempts have been made to model the effect of relative permeability on instabilities [17–20]. As Riaz and Tchelepi [17] reported, viscous fingering characteristics of two-phase flows are strongly dependent on the relative permeability profiles.

Lenormand [10] conducted displacement experiments in micromodels that spanned a range of *Ca* and *v*. He concluded that three main displacement patterns are achieved during immiscible displacement: (1) viscous fingering, (2) capillary fingering and (3) stable displacement. For v > 1 and at low *Ca* (10⁻⁹), the displacement is in the form of capillary fingering; however, for larger *v*, the displacement becomes more stable. Conversely, for v < 1 and at high *Ca* (10⁻⁴–10), viscous fingering dominates the flow, due to the lower viscosity of the nonwetting liquid, while at low *Ca* (10⁻⁹), capillary fingering can be

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https://doi.org/10.1016/j.fuel.2018.08.086

Received 23 April 2018; Received in revised form 3 July 2018; Accepted 21 August 2018 0016-2361/@ 2018 Elsevier Ltd. All rights reserved.





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observed. Viscous fingering during immiscible displacement in porous media can result in early water breakthrough, which would lower the efficiency of oil recovery [21,22]. Both viscous and capillary fingering can be observed at moderate Ca (10^{-4} – 10^{-6}) with v < 1, typically termed as the crossover zone. As proposed from Lenormand [10], a phase diagram can be generated to parametrise the expected flow regime, in terms of Ca and v.

Many research groups have investigated displacement patterns using micromodels and various surfactant cocktails [23-28]. Yadali Jamaloei and Kharrat [29] conducted a study of two-phase flow using a dilute surfactant solution in both oil-wet and water-wet micromodels. Their results in the oil-wet micromodel showed formation of water-inoil microemulsion, and formation of partially continuous surfactant solution. The water-wet micromodel showed inter-pore bridging, deformation and stringing of the residual oil, which enhanced oil recovery. Dong et al. [30] studied the displacement mechanisms of alkaline flooding for heavy oil in glass etched micromodels. Two displacement patterns were observed: (1) water-in-oil (W/O) microemulsion formation and partial wettability alteration; and (2) oil-inwater (O/W) microemulsion formation, resulting in dispersion of the heavy oil in the water phase. Other than typical surfactant floods, microemulsion-based oil recovery has received much attention in the past decade [5,31], due to the ultra-low IFT and higher viscosity that can be achieved [32,33]. In addition, several researchers have conducted coreflooding experiments to test the effectiveness of microemulsion flooding [33-37]. Overall, all of the surfactant-based microfluidic studies have observed deformation of fluid/fluid interfaces and/or formation of emulsions that are indicative of near miscible conditions [23-28].

The basic mechanism of microemulsion formation is generally understood with the aid of phase behaviour tests [1]. These tests provide the equilibrium arrangement of components within two or more phases and are necessary to examine the suitability of an EOR technology for a given field application [1,38,39]. In particular, surfactant-oil-brine phase behaviour is sensitive to changes in brine salinity. Tests are often conducted to evaluate a pre-selected range of different salinities [1]. Equal amounts of oil and EOR fluid at a particular salinity can be added to a tube and mixed vigorously and then left stagnant to separate [1,3]. During equilibration, components (oil, water and surfactant) separate into distinguishable phases of which unique phase properties are often characterised by the amount of component that constitutes a given phase. For example, the Huh [40] equation uses the solubility ratios of water-to-emulsion (γ_w) and oil-to-emulsion (γ_o) to determine interfacial tension (IFT). Salinity plays a major role in the amount of component that partitions to a given phase and thus, for a pre-selected range of salinities different Winsor phase behaviours occur. Winsor type IIphase behaviour is when $\gamma_w > \gamma_o,$ resulting in most of the microemulsion to appear in the brine. Conversely, Winsor type II+ corresponds to when $\gamma_w < \gamma_o$, therefore, the microemulsion is mostly in the oil phase. Winsor type III occurs when γ_w equals $\gamma_o,$ which creates a microemulsion between the oil and brine phases [3,5,38,39].

Herein, we aim to characterise displacement processes that occur during surfactant flooding by using a microfluidic approach. We use a surfactant solution where the components provide a range of different phase behaviours depending on brine salinity. We explore how phase behaviour influences the resulting displacement patterns under equivalent flow rates. Pore-scale displacement studies have shown that phase behaviour can significantly influence fluid flow within a single pore [8] and we extend this work by studying fluid/fluid displacement over multiple pores in a simple microfluidic device.

2. Materials and methods

2.1. Chemicals

Enordet J13131 was selected as the surfactant for this study. It is an

Table	1			

Details	or	the	chemicals	usea	ın	the	study.

Chemical	Provider	Commercial name/code
Sulfonated HPAM	SNF	P-071006
Alkyl ether sulphate	Shell	Enordet J13131
Co-solvent	Sigma-Aldrich	2-Butanol
Water soluble dye	Sigma-Aldrich	Fluorescein sodium salt

alkyl ether sulfate surfactant with a distribution of 12–13 carbon atoms produced by Shell Chemicals, which is a part of the ENORDET O series. Sulfonated hydrolysed poly-acrylamide (HPAM) by SNF chemicals was chosen as the polymer. HPAM comprises of acrylamide monomers, a fraction of which have been hydrolysed. Both surfactant and polymer were chosen due to their relevance to EOR field projects [8,41]. Details of the chemicals are provided in Table 1 and the basic chemical structures are provided in Fig. 1.

2.2. Static equilibrium phase behaviour test

Surfactant-oil-water phase behaviour tests for a range of salinities (4.50-5.75% sodium chloride (NaCl) (w/v)) were prepared with equal volumes of water and oil. Aqueous solutions of surfactant with co-solvent and salt were prepared as listed in Table 2. Fluorescein sodium salt (Sigma Aldrich, USA) was used in the brine phase for visualisation purposes during microfluidic experiments. Therefore, all of the phase behaviour tests and microfluidic experiments were conducted with the fluorescent dye. The solubilisation ratios were analysed for a range of salinities, as previously described in [24]. Changes in the brine salinity displayed different Winsor type behaviour (type II – , II + and III). We then select one salinity for each Winsor type behaviour for microfluidic experiments. Winsor type III behaviour was selected based on equal oil and water solubilisation ratios for the microemulsion phase [8]. Winsor type II- (oil-in-water emulsion) was selected when the volume of water in the microemulsion is greater than the volume of oil. Conversely, Winsor type II+ (water-in-oil emulsion) was selected when the volume of water in the microemulsion is less than the volume of oil.

2.3. Dynamic phase behaviour test

Surfactant–oil–water phase behaviour tests at the three different salinities that represented each Winsor type behaviour were selected for dynamic testing. The test tubes were shaken vigorously and left standing vertically and imaged continuously using a digital camera (Basler acA1600-60gc GigE camera, Basler, Germany). One image was taken every 10 min, and the rate of microemulsion formation was measured from the digital images for 24 h.

2.4. Interfacial tension and rheology measurements

Interfacial tension (IFT) between microemulsion/oil (σ_{mo}) and microemulsion/brine (σ_{mw}) were estimated using the Huh [40] equation, which relates IFT to the solubilisation parameters:

$$\sigma_{mi} = \frac{0.3}{\gamma_i^2} \tag{2}$$

where σ_{mi} is the IFT (in mN/m) between microemulsion and phase *i*, and γ_i is the corresponding solubilisation parameter. The coefficient of 0.3 corresponds to the microemulsion distribution as described in Huh [40], which is sensitive to the type of surfactant used. These results were then compared against IFT measurements obtained by a spinning drop tensiometer (M6500 Grace Instrument, United States). The sample holder was filled with surfactant solution at a given brine salinity (aqueous phase) and then a drop of microemulsion was placed at the top. Measurements were performed for at least 20 min at ambient

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