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Synergism, phase behaviour and characterization of ionic liquid-nonionic surfactant mixture in high salinity environment of oil reservoirs

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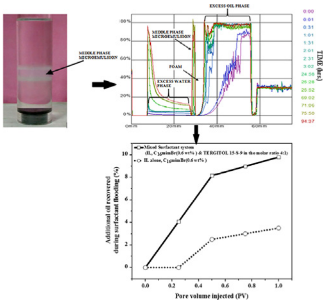
GRAPHICAL ABSTRACT



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ABSTRACT

A large number of matured oil reservoirs still have ample quantity of oil trapped in the pores of the reservoir rock. The quest of a reliable tertiary oil recovery process has opened opportunities for chemical enhanced oil recovery processes. Most of the conventional chemicals used in this method lose their efficacy under high salinity conditions of oil reservoirs. In the present work, synergism in reducing interfacial tension between crude oil and aqueous phase has been investigated, between a mixture of long chain imidazolium ionic liquid and a nonionic surfactant. The reservoir fluid has been modelled as a system containing crude oil/kerosene and surfactant brine. Phase behaviour studies of this system at varying salinities have been studied. Formation of a distinct and stable TYPE III microemulsion at ultra-high salinities has been observed. Presence of middle phase microemulsion at very high salinities indicated ultralow interfacial tensions thereby making the mixed surfactant formulation suitable enough to be used in reservoirs having high salinity of formation brine. Characterization of this middle phase microemulsion was carried out using Transmission Electron Microscopy (TEM), Atomic Force Microscopy (AFM), Polarised optical microscopy (POM) and Dynamic light scattering (DLS) techniques. Time required for the modelled reservoir fluid to coalesce into a stable middle phase microemulsion was characterised using Turbiscan. Subsequently, flooding experiments with only ionic liquid (C_{16}mimBr) and mixed system containing ionic liquid (C_{16}mimBr) + nonionic surfactant (Tergitol 15-S-9) at high salinity conditions were performed. The results showed that during surfactant flooding with C_{16}mimBr alone, the amount of oil recovered was 3.7% of the original oil in place whereas for C_{16}mimBr + nonionic surfactant mixture, percentage of oil recovered was 9.79% of the original oil in place.

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1. Introduction

Enhanced Oil Recovery (EOR) methods have been proposed to recover residual oil that is trapped in the pores of the oil reservoir after primary and secondary oil recovery processes. Surface active agents and/or alkali are used in chemical flooding method to reduce interfacial tension (IFT) between oil and water in the reservoir, whereas polymers are used to enhance displacement sweep efficiency [1]. During surfactant flooding, IFT between crude oil and reservoir water should be decreased to at least 10^{-2}–10^{-3} mN/m. Moreover, for high percentage of oil recovery such ultralow IFT should be maintained during the entire surfactant flooding process [1–5]. Chase water is then flushed into the reservoir which displaces the slug thus developing an oil bank towards the producer well. Petroleum sulfonates, anionic surfactants in nature, are used as surface active agents in EOR. Nonionic surfactants are preferably used as co-surfactants to increase the effectiveness of the primary surfactant in reducing the interfacial tension [1]. The most important criteria while selecting a surfactant for surfactant flooding process is that it should have the ability to lower the interfacial tension and retain its IFT under reservoir conditions of high salinity and high temperature thus breaking up the oil into microemulsion so that they can be extracted from the pores by hydrodynamic force of the chase water. High oil solubilisation rates, formation of low viscosity microemulsions and minimal loss of surfactant by adsorption on reservoir rocks are also some of the other important criteria [5]. However it is found that most of the conventional surfactants used in the EOR process lose their efficacy under harsh reservoir conditions (high salinity and high temperature). Zhang et al. [6] performed phase behaviour and IFT studies for six different surfactant formulations and crude oil (43.2° API) from the Bakken Formation. At high temperature (80–120 °C) and high salinity (15–30% TDS) conditions of Bakken reservoirs, the IFT between crude oil and surfactant brine liquids increased with increasing temperature and salinity. Also the most efficient surfactant had an optimal salinity of (4–6% TDS), which is very low for such high saline reservoirs [6]. In another work, Aoudia et al. [7] also carried out studies of oil displacement from cores at high temperature and salinity. The brine solution used in the study was formation brine from Yibal oilfield in Oman having an extreme saline environment (20 wt%). Most of the conventional surfactants tested for the reduction of IFT in such reservoirs lost their efficacy under the harsh saline condition [7]. These drawbacks demand a more extensive research for developing environmental friendly surfactants which can sustain such high salinity conditions and at the same time exhibit ultralow interfacial tensions.

Organic salts having melting temperature below 100 °C are known as Ionic Liquids (ILs). Unlike conventional organic salts, ILs possesses high ionic conductivity, high thermal stability, high solvating capacity and wide liquidus range. Since ionic liquids have negligible vapour pressure they are less toxic than the conventional organic solvents and are thus referred as environmental friendly “Green solvents”. In addition cations and anions of ILs can be easily tuned into make task-specific solvents [8–10]. Many ILs are amphiphilic substances having hydrophilic head and hydrophobic tail, exhibit surface activity and have ability to self-aggregate. Reported literatures conclude that imidazolium based ionic liquids behave like conventional cationic surfactants [11,12]. Micellization in solutions where ILs act as surface active solutes is studied intensively since decades and it is found that ILs can replace conventional surfactants in fields of drug delivery, nanosynthesis, synthesis of porous silica materials etc. [11–13].

In the past few years researchers have studied the behaviour of IFT reduction between crude oil/water in the presence of ILs [14–23]. Heazve et al. [17] have studied the effect of different imidazolium based and pyridinium based ionic liquid surfactants on the IFT between crude oil and water. They found that interfacial tension between crude oil and IL-brine solution decreased even under high salinity (~ 30 wt%) and high temperature (~ 60 °C) conditions. The imidazolium class of ionic liquids were more effective in reducing IFT than pyridinium type of ionic liquids of same alkyl chain length. In another study Lago et al. [18] studied the phase behaviour of water/brine – ionic liquid trihexyl (tetradecyl)phosphonium chloride – dodecane. The phase equilibrium study indicated the formation of a Winsor type III system, with a middle phase microemulsion. In our previous study, long chain imidazolium ionic liquid, C_{16}mimBr was deployed as a surfactant in the process of surfactant flooding during tertiary oil recovery process from an oil filled sand-pack reservoir core. The percentage oil recovered by C_{16}mimBr during the EOR process was then compared to that recovered when using a conventional cationic surfactant, Cetyl trimethylammonium bromide (CTAB) [23].

All the previous works, investigating the use of individual ionic liquids as surfactants for chemical EOR processes (surfactant flooding) suggest them as potential candidates in reducing IFT. However, the obtained interfacial tensions are very high (> 1mN/m) and do not fulfil the insistent demand of ultra-low values of IFT [14–17]. A few researchers have studied about the effect of blends of surface active ionic liquids and common anionic surfactants in reducing IFT between oil and brine. Jia et al. [24] have reported that a mixed cationic surface-active ionic liquid/anionic surfactant system consisting of N-dodecyl-N-methylpyrrolidinium bromide and sodium dodecyl sulfate reduced the IFT between water and oil, and the intensity of the effect depended on the cationic/anionic surfactant ratio. [24]. In another study Iria Rodriguez-Escontrela et al. [25] found that two anionic and two cationic, surface active ionic liquids individually have little potential for EOR but blends of an anionic internal olefin sulfonate surfactant with cationic surface active ionic liquids reduced IFT between n-octane and aqueous solutions to ultralow values at an optimal blend ratio [25]. Hence in the present work, in order to fulfil the inevitable demand of ultralow IFT between crude oil and ionic liquid surfactant slug, ionic liquid – nonionic surfactant mixtures have been used as surfactant slugs. In the present work, mixture of ionic liquid, 1-hexadecyl-3-methyl imidazolium bromide (C_{16}mimBr) and a nonionic surfactant, TERGITOL 15-S-9 is used as a surfactant solution for the purpose of recovering residual oil from artificial cores. The main objective of this study is to investigate synergism/antagonism between the two surfactants in reducing interfacial tension between crude oil-aqueous solutions. Until now scientists have tried to find out the potential of long chain ionic liquids as EOR surfactants however no studies based on synergism between ionic liquid-nonionic mixtures in reducing IFT and their application in EOR processes has yet been reported. In order to determine the effectiveness of the new C_{16}mimBr – TERGITOL 15-S-9 surfactant mixture in a proposed surfactant flooding EOR method, phase behaviour studies at ultra-high salinity conditions have been performed. The effectiveness of the newly proposed surfactant mixture (C_{16}mimBr-TERGITOL 15-S-9) in recovering residual oil during surfactant flooding of the core has also been studied.

2. Materials and methods

2.1. Materials

In the present work 1-hexadecyl-3-methyl imidazolium bromide (C_{16}mimBr) was prepared and purified by the procedure mentioned elsewhere [26]. In our previous work, surface tension was measured to determine the critical micelle concentration (CMC) of 1-hexadecyl-3-methyl imidazolium bromide (C_{16}mimBr) [23]. Absence of a minimum near the CMC confirmed the purity of the surface active ionic liquid, C_{16}mimBr. The nonionic surfactant, TERGITOL 15-S-9 (99% purity), was purchased from Sigma-Aldrich. Sodium chloride was purchased from Finar Chemicals. Commercially available purified kerosene was purchased from the local market. Kerosene is a crude oil distillate and is a mixture of branched and straight chained with no definite molecular formula. An FTIR spectra of kerosene that has been used in this study is provided in the Fig. 1(a). The infrared spectra of kerosene shows absorption peaks at 2925.15 cm^{-1}, 1462.09 cm^{-1} and 1377.22 cm^{-1} in