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Adsorption of aliphatic ionic liquids at low waxy crude oil–water interfaces and the effect of brine

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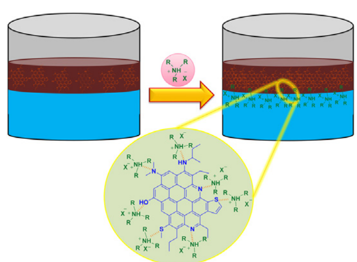
HIGHLIGHTS

- Aliphatic ionic liquids reduce the interfacial tension of crude oil–water system.
- Ionic liquid + NaCl for crude oil–water system shows synergism in surface phenomena.
- Ionic liquid with $[HSO_4]^-$ or $[H_2PO_4]^-$ anion shows better efficiency.
- Greater the chain length of ionic liquid, higher is the tendency to lower surface/interfacial tension.
- Applicable for the recovery of residual oil from matured reservoirs.

GRAPHICAL ABSTRACT

Proposed mechanism showing the effect of aliphatic ILs in decreasing the IFT of crude oil–water system.



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ABSTRACT

The need for development of better surface active agents for upstream oil and gas industry which can survive harsh condition of salinity are in great demand, particularly for the applications related to improved/enhanced oil recovery, flow assurance and oil and gas production operations keeping in mind the environmental constraints. The technical difficulties which need to be considered are those involving the surface forces such as surface tension (SFT) and interfacial tension (IFT) acting between the formation water and the low waxy crude oil. In this study, we have employed the use of *eight* aliphatic ionic liquids (ILs), based on di- and tri-alkyl ammonium as cations and with various anions such as $[CH_3COO]^-$, $[BF_4]^-$, $[H_2PO_4]^-$ and $[HSO_4]^-$ for the investigation of the surface phenomenon of crude oil–water system. The synergistic effect of NaCl along with the ILs is investigated in detail. It is observed that there is a significant reduction in the surface tension of water and the interfacial tension of crude oil–water system in the presence of salt, particularly at higher concentration of NaCl (200,000 ppm). Effect of temperature, time, alkyl chain length of the cationic part of the ILs, nature of anions of ILs and the concentration of ILs is also discussed. The trend in the electrical conductivity of aqueous IL solutions with various concentrations at three different temperatures 298.15–318.15 K is also presented along with critical aggregation concentration. The study on the effect of ILs on the SFT/IFT of water and low waxy crude oil–water system reveal that the ILs are successful in minimizing the effect of the surface forces in the presence of salt and thereby, could pave the way for efficient enhanced oil recovery operations.

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1. Introduction

Interfacial science has a dynamic importance in the field of petroleum engineering, especially, when it comes to the recovery of unconventional crude oil and the residual oil trapped in reservoirs. Recovery of residual oil trapped inside the reservoir and enhanced oil recovery are mostly influenced by key factors namely, capillary forces, viscous forces, contact angle, wettability, surface tension (SFT) and interfacial tension (IFT). Reduction in the capillary forces and interfacial forces helps in smoothening the mobilization of residual oil followed by its recovery. Mobilization facilitation of residual oil requires a reduction in the capillary forces within the oil reservoir which is to be achieved by lowering the IFT during flooding processes [1]. Furthermore, the high salinity conditions of the reservoir make it difficult to reduce the interfacial tension in the typical way. Classical methods such as, polymer flooding and surfactant flooding lose their efficiency at high salinity and high temperature conditions.

Throughout our biosphere, trapped oil in depleted reservoirs count for around two-thirds of the total crude oil reserves, remaining unrecovered in the reservoirs, even after primary and secondary recovery stages [2]. Several studies were carried out in the past, so as to improvise oil recovery techniques and, thereby, enhance oil production [3,4]. Oil and gas industries, throughout the globe are in the quest of acquiring well developed, efficient and economically friendly techniques for forcing out the oil which is trapped in the oil reservoirs [5–8]. It is at this stage, researchers tried to develop new strategies for employing ionic liquids for improved enhanced oil recovery (EOR) techniques. Though much attention on this aspect prevails, at present there is only inadequate information on physical properties known for these systems with respect to crude oil for successful engineering applications [9].

Ionic liquids (ILs), widely known as green solvents [10–13], are organic salts possessing very low melting points [14]. ILs possess good catalytic properties, negligible vapour pressure, non-flammability, high chemical and thermal stability, etc. Over the last decade, ILs have been broadly used for a wide range of applications in science and engineering, such as organic and inorganic synthesis, extraction and separation, catalysis, electrochemistry, biomass conversion, etc. [15–18]. The main reason behind their extensive classes of application is due to their flexible behaviour, which can be tuned systematically by varying the cationic and anionic components. The ionic environment created by ILs enables various types of interactions to be possible. Details regarding the physicochemical properties of ILs become mandatory not only for process design but also for the development of comparisons and estimation of these properties [19–21].

Many researchers [22–24] have carried out investigations on the IFT between two immiscible liquids using various methods, such as the Wilhelmy plate, Du Noüy ring, drop volume, sessile drop, spinning drop, pendant drop, Neumann triangle, imbedded fibre retraction, breaking thread method, etc. The selection of the method depends on various factors such as, properties of the liquid under study, the circumstances in which the experiment is carried out and the stability of the liquid. The author Hezave et al. [6–8] observed significant reduction in the interfacial tension of crude oil/water system using the ionic liquids, such as 1-dodecyl-3-methylimidazolium chloride ([C_2-mim][Cl]), 1-octyl-3-methylimidazolium chloride ([C_8-mim][Cl]), 1-dodecyl pyridinium chloride ([C_{12}-Py][Cl]) and 1-octyl pyridinium chloride ([C_8-Py][Cl]). The same ILs were tested at high saline conditions as well, and inferred that, these ionic liquids efficiently helped in decreasing the interfacial tension. Ultimately, it was found that the IFT measurements had a correlation with salinity and temperature. 1-Dodecyl-3-methylimidazolium chloride ([C_2-mim][Cl]) was projected as a better agent in lowering the IFT and the critical micelle concentration (CMC) between crude oil and brine (NaCl). The CMC was obtained at a lower concentration of 100 ppm [6].

Wang et al. [25] investigated the IFT of a few [C_n-mim][BF_4]/alkane systems and [C_n-mim][PF_6]/alkane systems at different temperature range (288.15–328.15 K) at atmospheric pressure. It was detected that on increasing the temperature of the system, IFT decreased linearly. Similar results were observed when increasing the alkyl chain length of head group of ionic liquids. The same trend was observed for [C_n-mim][PF_6]/alkane system. The information on the IFT of a few [C_n-mim][BF_4]/alkane systems and [C_n-mim][PF_6]/alkane systems were measured by Wang and his co-workers. The interfacial tensions of the four imidazolium based ILs ([C_n-mim][BF_4]; where n = 3–6) with hexane, heptane, and cyclohexane were taken at different experimental temperatures in the range of 288.15–328.15 K at atmospheric pressure. It was identified that, with increase in temperature, the interfacial tension decreased. The interfacial tension values were also found to decrease with increase in alkyl chain length for all the ILs/alkane system. A linear decline of IFT with rise in temperature was also observed. Similar results were observed for the interfacial tensions of four other imidazolium based ILs ([C_n-mim][PF_6]; where n = 5–8) with hexane and heptane [25,26].

Gardas et al. [27] measured the interfacial tension of water and several *n*-alkane system (*n*-hexane/*n*-octane/*n*-decane) in the presence of various imidazolium ionic liquids in the temperature range of 293–323 K with an interval of 10 K at atmospheric pressure. They found that the ionic liquid containing longer chain length on the header part showed a better performance on the reduction of interfacial tension of the system. On the other hand, they also observed a decreasing trend on the IFT on increasing the temperature. In addition, they also noted that the IFT of any ILs/*n*-alkane system was much lower than the individual IFT of ILs in air, and IFT of *n*-alkane in air.

The classical surfactants were not found to be effective as compared to ILs in reducing the IFT at high temperature conditions. Ye et al. [14] performed the IFT analysis of crude oil/water system by using gemini surfactants, such as butyl-α, ω-bis (tetradecyldimethylammonium bromide) and the butyl-α, ω-bis (hexadecyldimethylammonium bromide). In this case, the author noticed that an increase in the temperature of the system in the presence of surfactants did not improve the reduction in IFT as expected. However, for the study of the effect of temperature, combination of surfactant system was not found to exhibit significant effect on the IFT due to synergism [14]. It is, thus, inferred that the ILs are capable to decrease the IFT considerably, that is otherwise, quite impossible for the conventional surfactant especially at high salinity and high temperature conditions. It was also found that the effect of ionic liquid on IFT reduction was encouraging and in addition, higher concentration of salt did not affect the stability of the IL solution [6–8].

Effectual separation of any liquid from a mixture of immiscible liquids primarily involves the phenomenon of SFT and IFT being alleged as the dependent factors. The interfacial tension between bitumen/silica systems was found to be decreased in the addition of ionic liquids, facilitating the easier recovery of bitumen [28]. In the case of bitumen and silica, the adhesion force is decreased in the presence ionic liquids than in the normal aqueous solution. Similarly, the contact angle of bitumen/water droplet was about ∼90°, but in the case of bitumen/ionic liquids system it decreased to an angle of ∼73° [29]. These results illustrate that the separation is easier by using aqueous solution of ionic liquids rather than only with the water.

Vast unmapped space in the field of crude oil recovery demands for crafting and employing suitable ionic liquids. Even though numerous investigations [6–8,10,11,30] have been performed in this respect, it is found that the majority of the conventional

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