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# Application of different novel and newly designed commercial ionic liquids and surfactants for more oil recovery from an Iranian oil field



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#### ABSTRACT

This investigation is conducted on one of the southern Iranian oil fields, which experiences a fault on its gas cap. Therefore, no traditional gas injection and normal water injection for pressure maintenance is suggested. The target is set to inject special compatible and proper type of chemicals in each single well to avoid the pressure increase for the risk of oil and gas spill. Two different sources of aqueous phases, namely formation brine and sea water were used to find the effects of different brine composition on the main mechanisms of releasing the trapped oil, interfacial tension (IFT) and wettability alteration. The obtained results reveal that both the ionic liquid (IL) based surfactants and new synthesized commercial surfactants are not only able to tolerate harsh conditions (250,000 ppm) of salinity but also have the ability to reduce the IFT more effectively (from 19 to 0.07 mN m<sup>-1</sup>). As the salinity is increased, ionic liquid ([ $C_{12}mim$ ] [CI]) showed lower critical micelle concentration (CMC) of about 50 ppm, which shows that the IL use for EOR is economically feasible. The contact angle measurements also indicate the possibility of activating wettability alteration mechanism.

Consequently, the core flooding tests revealed more oil recovery efficiency of 8% and 22% based on original oil in place (OOIP) and residual oil in place, respectively for the solely IFT reduction cases. The oil recovery efficiency for the tertiary oil recovery process was increase up to 12% and 35%, based on OOIP and residual oil in place, respectively, which clearly clarifies the effectiveness of soaking to exploit the wettability alteration mechanism.

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#### 1. Introduction

Due to a decrease in oil prices over the last couple of years, low cost and effective enhanced oil recovery (EOR) techniques have become more popular which in turn has inevitably lead to an increase in crude oil consumption [1]. Additionally, researchers have been motivated to develop more efficient secondary and tertiary enhanced oil recovery methods for the past decades for oil recovery techniques to be more economical.

Among different possible processes for tertiary oil recovery, chemical enhanced oil recovery (CEOR) methods are more appealing as they are very effective if the best and most optimized chemical is chosen. The surfactants, as the most well-known chemical agent for EOR, are identified to activate two main mechanisms including IFT reduction and wettability alteration in the oil reservoirs [2–6]. Wettability alteration affects the distribution of water and oil in the rock/fluid system while IFT reduction directly affects the fluid-fluid interface forces and facilitates the movement of trapped oil [7]. Since the capillary force is directly related to the rock's wettability and the IFT of oil and water phases, it is possible to consider their variations as the indication of oil recovery efficiency using chemicals such as surfactants [8]. The recent progress to design targeted-molecule chemicals has enabled the researchers to propose new surfactants which are not only capable to tolerate harsh conditions of oil reservoir, such as salinity and temperature, but also have the capability to introduce lower adsorption rate on rock while the IFT reduction is more profound [9–16].

Among the different parameters affecting IFT of crude oil/water systems the salinity, especially the presence of different mono and divalent ions [17], is the most important one [18–21]. The most recent published works reveal that the type of ions and their concentration in the brine as well as the oil composition have significant effects on the IFT variation for different types of crude oils and saline solutions [15–17,22–25].

Besides, different types of active agents were proposed as IFT reducing agents in the past decades to be used as chemical enhanced oil recovery (CEOR) agents. Ionic Liquids (ILs) and functionalized surfactant molecules have gained attentions during the recent years because of their unique

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advantages. In details, ILs are defined as salts which are in liquid form at ambient or even far below ambient temperature. Since ILs are comprised entirely of ions, they have very unique advantages making them appropriate candidates for multidisciplinary purposes [26] especially in the oil industry. They can be used to stabilize the asphaletene and resin content of crude oil, as well as desalting and demulsification of water-in-crude oil emulsions. Painter et al. reported that up to 90% of extra-heavy oil recovery from Canadian tar sands using common ILs like [Bmim]<sup>+</sup>[CF<sub>3</sub>SO<sub>3</sub>]<sup>-</sup> and [Bmim]<sup>+</sup>[BF<sub>4</sub>]<sup>-</sup> with five times recycling process without noticeable loss of efficiency [27,28].

Regardless of wettability measurement methods, the type of surfactants (anionic or cationic) may introduce different trends on wettability alteration. For example, in 1997, Austad and Milter observed that cationic surfactants are more effective compared to anionic surfactants to change the wettability of low permeable chalk toward more water-wet state. They associated their observation to the fact that cationic surfactants lead to the formation of ion pairs between cationic heads of surfactant molecules and acidic components of the crude oil adsorbed on the surface of carbonate rock. However, molecules of ionic surfactants could form a monolayer on the rock surface through hydrophobic interactions with the adsorbed crude oil components. The layer of the adsorbed surfactants with the hydrophilic head groups covering the originally oil-wet rock could change the wetting state of the rock toward more water-wet state [29–31].

This especial experimental study has been conducted on one of the southern Iranian oil fields, which experiences a fault on its gas cap. As a result, performing traditional injection processes, like gas and normal water injection for pressure maintenance is not proposed. The most affordable proposed target is to inject special compatible type of chemicals in each single well to stimulate them to avoid the increase in pressure for the risk of spill from the reservoir area. One of the main targets of this investigation is to synthesize, new type of surfactants for both, effectively reducing the IFT and wettability changes as well as tolerating the harsh conditions of salinity up to 250,000 ppm. To maintain more real conditions in the experiments, the original rocks, brine and crude oil from the oil reservoir have been utilized. As far as the researchers of this paper could find, there is no report on the application of surfactants proposed in the current study for IFT reduction and wettability alteration of a crude oil from this southern Iranian oil fields at high salinity conditions using available resources of water including, formation brine, sea water and distilled water.

In this regard, emulsification and compatibility tests were performed at the first stage to select proper surfactants which could tolerate harsh conditions of salinity. Then, the IFT of compatible solutions introduced the best emulsification behavior which was measured to determine the effect of salinity on IFT. Subsequently, contact angle measurements were performed to find the possible effect of these surfactants on the wettability alteration. Finally, several coreflooding tests with different injection patterns including the quick coreflooding process (alternative injection without any soaking during the injection process) and coreflooding process followed by soaking period (21 days) were performed to find the effectiveness of the screened chemicals on the tertiary oil recovery for possible field application.

#### 2. Experimental section

#### 2.1. Materials

#### 2.1.1. Ionic liquids

The supplied 1-methylimidazole, 1-chlorododecyl and diethyl ether from Merck/Fluka were used to synthesis the desired ILs as previously described in details elsewhere. <sup>36</sup> In brief, the 1-methylimidazolium come into contact with an excess of the 1-chlorododecane without additional solvent in around-bottomed flask fitted with a reflux condenser (with heating and stirring at 70 °C for 48–72 h) to synthesize 1-dodecyl-3methylimidazolium chloride ([ $C_{12}$ mim][Cl]) and 1-Octadecyl-3-methylimidazolium chloride ([ $C_{18}$ mim][Cl]). In this stage, the produced viscous liquid is kept at ambient condition to cool down, and then rinsed with diethyl ether. Then, the remained substance is dried at 100 °C overnight and in the next phase; its purity is examined using H-NMR spectroscopy. Sonication technique was used to disperse and dissolve these IL-based surfactants into the aqueous medium.

#### 2.1.2. Commercial surfactants

Five different new surfactants namely AN-120, NX-610, NX-1510, NX-2760 and TR-880 were designed and synthesized to examine their effectiveness tertiary oil recovery. (See as listed in Table 1)

#### 2.1.3. Crude oil and aqueous media

The crude oil used in this investigation was obtained from one of the southern Iranian oil fields close the Persian Gulf with an API<sup>o</sup> of 26. In addition, three different aqueous phases, namely reservoir brine (ppm) (Na<sup>+</sup>&K<sup>+</sup>:88,568,Ca<sup>2+</sup>:10,160, Mg<sup>2+</sup>: 1798, Cl<sup>-</sup>:133,835, SO<sub>4</sub><sup>2-</sup>: 1104, HCO<sub>3</sub><sup>-</sup>: 439), sea (Persian Gulf) water (K<sup>+</sup>: 501, Ca<sup>2+</sup>: 478, Mg<sup>2+</sup>: 1626, Cl<sup>-</sup>: 23,890, Na<sup>+</sup>: 13,810, SO<sub>4</sub><sup>2-</sup>: 3029) and deionized water, were used to find the effects of different available sources for possible future injections.

#### 2.2. Experimental apparatuses

#### 2.2.1. Interfacial tension and contact angle measurements

Since the IFT range to be measured in this study is in the order of  $10^{-2}$  to 50 mN m<sup>-1</sup>, both pendant and spinning drop methods were utilized for accurate measurements. For solutions with the IFT values between 0.1 and 72 mN m<sup>-1</sup>, a drop shape analysis (DSA 100, KRUSS, Germany) was used. Additionally, due to the advantage of drop shape analysis this method was also used to measure the equilibrium contact angle of crude oil/aqueous solution on the carbonate rock at the ambient pressure and temperature. The DSA 100 instrument was explained in more details in literature previously published by the same co-authors [1,32–34]. For IFT measurements, the crude oil droplet was formed at the tip of a needle and then the image of the pendant crude oil drop in aqueous solution was analyzed to measure IFT. In addition, contact angle was measured with sessile drop method.

Moreover, the IFT of solutions lower than 0.1 mN m<sup>-1</sup> were measured using spinning drop tensiometer (SITE-100; KRUSS, Germany). A detailed description of the used spinning drop has been given elsewhere [16].

#### 2.2.2. Emulsification and compatibility

In the first step, the compatibility of all the chemical agents dissolved in different types of water, was examined to find the proper chemical agents concentrations. In this regard, different solutions of chemicals with desired concentrations were prepared using distilled and sea water and formation

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