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Article

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Research paper

An experimental investigation into enhancing oil recovery using smart water combined with anionic and cationic surfactants in carbonate reservoir

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ABSTRACT

Among all EOR methods, chemical flooding using wettability alteration materials may be a promising choice to enhance oil recovery. The present study involves a combination of smart water and anionic and cationic surfactant injection as a new approach of EOR in an oil wet carbonate reservoir. CTAB as a cationic and SDS as an anionic surfactant were considered. The modified seawater from Persian Gulf was selected as the smart water. The effects of the smart water salt content and the concentration of the surfactants were measured. The optimum salt concentration of smart water was obtained when SW-3KCl, SW-1NaCl, and SW-4Na_2So_4 were used. Moreover, the optimum smart water combined with the surfactants was estimated to be 0.1CMC of SDS and 0.9 CMC of CTAB. The results showed that the combination of smart water and surfactants in carbonate reservoirs changed the wettability towards a water wet system. The oil recovery factor for distilled water, the seawater and the combination of smart water and the optimum surfactant concentrations were calculated to be 28%, 42% and 58%, respectively. The combination of smart water and surfactant had more oil recovery compared to an individual surfactant or smart water flooding.

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1. Introduction

Therefore, producing oil from oil reservoir is of paramount importance for the purpose of supplying the world's energy demands. Oil production from petroleum reservoirs is categorized into three main stages, including primary, secondary and tertiary recovery. It is well-known that during the first stage of the recovery, primary recovery, the natural reservoir pressure is responsible for sweeping oil towards the production wells. However, after a while, the reservoir depletes and the reservoir pressure declines with it, and therefore the natural capacity of reservoir to produce oil decreases (Ramachandran et al., 2003). The main purpose in the secondary recovery stage is to maintain the reservoir pressure by injecting fluids, such as water and gas. Water and gas injection with the purpose of pressure maintenance are effective when the water cut and gas oil ratio of the reservoir are not significant (Planckaert, 2005; Saeedi Dehaghani and Rahimi, 2018). In the third stage, Enhanced Oil Recovery (EOR) methods are employed to recover the oil that was not produced during the first two stages of recovery (Gbadamosi et al., 2019). EOR methods are divided into four main groups, including thermal, miscible/immiscible gas injection, microbial, and chemical methods (Goodlett et al., 1986). Among the four aforementioned methods, chemical flooding is the most suitable choice for reservoirs containing light oil. Chemical enhanced oil recovery (CEOR) methods involve mobility control of the injected fluid using polymer, wettability alteration and interfacial tension(IFT) reduction using surfactant, combination of polymer and surfactant, and foam flooding (Levitt and Pope, 2008; Sun et al., 2017; Samanta et al., 2012; Shang et al., 2019; Ding and Rahman, 2017). Rock and fluid properties and interactions are among the most important criteria in selecting an EOR method (Goodlett et al., 1986).

Among the aforementioned CEOR methods, wettability alteration and IFT reduction methods are utilized to improve oil recovery in oil-wet reservoirs. In recent years, advanced water flooding has received a great attention in the industry (Su et al., 2018; Al-Saedi and Flori, 2019). Adding a few types of ion to the injection water can cause the overall oil recovery to increase. Doing this, the presence of bivalent metallic cations (Ca^{2+}, Mg^{2+}) and So_4^{2-} may alter the wettability of the rock from oil-wet to water-wet in carbonate rocks. Therefore, selecting the optimum concentrations of these ions is of paramount importance, and hence by optimizing their concentrations, more oil may be produced from carbonate reservoirs (Fathi et al., 2010; Park et al., 2018;

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Dehaghani and Daneshfar, 2019; Duffy et al., 2019). Injecting the water with the optimized salt concentrations is named smart water flooding.

Moreover, surfactants can be used to lower the interfacial tension between oil and water. Surfactants (surface active agents) are organic complexes with at least one lyophilic and one lyophobic group in their molecule (Seethepalli et al., 2004b,a; Pal et al., 2018). Oil recovery from porous media can be improved using surfactants. In a reservoir, oil recovery strongly depends on the IFT and the capillary force (Stegemeier, 1977). As shown in Eq. (1), capillary number depends on IFT (Mohsenatabar Firozjaii et al., 2018; Dong et al., 2010). On the other hand, residual oil saturation depends on the capillary number. By increasing the capillary number, the residual oil saturation in porous media decreases, too. Since surfactants can reduce the IFT between oil and water, their injection to the reservoirs can cause the ultimate residual oil saturation to decrease, hence increasing the oil recovery.

$$N_c = \frac{u \times \mu}{\delta} \tag{1}$$

where u is velocity, μ is viscosity, and δ is IFT.

The most significant IFT reduction occurs at the critical micelle concentration of the surfactants (CMC). At CMC, the surfactant monomers existing in the emulsion have the highest possible concentration, thus reducing the IFT the most (Li et al., 2016). The hydrophilic portion of a surfactant may carry a positive or negative charge, both positive and negative charges or no charge at all. These are classified as cationic, anionic, amphoteric or non-ionic, respectively (Rosen and Kunjappu, 2012).

Researches have been conducted on using modified seawater as smart water (Hiorth et al., 2010; Strand et al., 2008, 2006; Webb et al., 2005). For example, Webb et al. (2005) conducted a comparative study on oil recovery from a carbonate core at reservoir conditions by injecting seawater containing $SO4^{2-}$ and simulated $SO4^{2-}$-free brine. It was determined that the wettability alteration of the carbonate rock with $SO4^{2-}$ ion is responsible for the saturation changes.

In the present study, oil recovery from a carbonate oil-wet micromodel using brine flooding, smart water flooding, and combination of smart water and surfactant flooding are investigated, experimentally. Modified Persian Gulf water is used as the smart water in this study. In addition, SDS and CTAB were the surfactants used in this study. By conducting experiments on the effects of the concentration of different salts on contact angle, the modified smart water with the optimum salt concentrations was selected. Then, SDS and CTAB were separately added to the optimum smart water to investigate their individual effects on contact angle. Then, the combination of SDS and CTAB in optimum smart water is prepared to obtain the optimum concentration of mixed SDS and CTAB in optimum smart water. Finally, brine, seawater, and optimum smart water mixed with SDS and CTAB are considered for flooding in a micromodel to determine oil recovery factor after each flood.

2. Experimental methodology

2.1. Materials

The crude oil used in this study was provided from one of the Iranian oil reservoirs located south west Iran. Properties of the crude oil are listed in Table 1. Composition of the formation water of the same reservoir was used to prepare a synthetic formation water. In order to prepare the synthetic formation water, the salts in the concentrations listed in Table 2 were used.

Table 1
Crude oil sample properties.

Gravity (API)	Asphaltene content (wt%)	Dead oil density (gr/mL)
27	5.72	0.893

Table 2
Formation water composition.

Salt name	Concentration (ppm)
NaCl	113 840
$BaCl_2$	318
$CaCl_2$	2560
$SrCl_2$	416

Synthetic seawater (in this case the Persian Gulf water) was chosen as the injection water used in this study. Table 3 lists the composition of the Persian Gulf water.

Surfactants

The two surfactants used in this study are: Cetyltrimethylammonium bromide known as CTAB and Sodium Dodecyl Sulphate known as SDS. CTAB is a cationic surfactant, while SDS is an anionic one (Llombart et al., 2019; Najafi et al., 2017; James-Smith et al., 2007).

2.2. Methodology

2.2.1. Aging

In order to conduct the contact angle experiments, 70 thin sections of the reservoir rock (1.5 cm*2.5 cm) were prepared and polished using sandpapers. Before being dried in an oven at 80 °C for an hour, the rocks were washed with water. In the next step, the clean and dry samples were immersed in Toluene so that their hydrocarbon contents are removed. The process continues until the Toluene stops changing color signifying that there is no more hydrocarbon residue in the rocks. Afterwards, the rock samples were put in an oven for twelve hours to dry out. Since there may still exist traces of different salts in the rocks, they were immersed in methanol for four days to allow the methanol to remove any traces of salts. Then, the samples were put in an oven again so that the alcohol evaporates. In order for the thin sections to be close to the actual reservoir conditions, they were immersed in formation water for seven days. At this point, a few tests were conducted to know whether the rocks have become water-wet or not. Once the samples were water-wet, they were aged in crude oil for four more weeks at 90 °C to allow them to become oil-wet.

2.2.2. Contact angle measurement

The sessile drop method was utilized to measure the contact angles. Since the oil is dropped on the surface of the rock in the presence of water, the higher the contact angle, the more oil-wet the rock is.

2.2.3. Glass-micromodel experiments

I this research, a heterogeneous five-spot glass-micromodel was utilized (Fig. 1). The model has an injection and a production well responsible for the diagonal flow through the porous medium. The model was made based on a thin section of rocks from carbonate reservoirs located south west Iran. Table 4 lists the properties of the model used in this study.

The micromodel flooding experiment steps are as follows:

1. The glass micromodel is saturated with a solution (95% Toluene + 5% Silane) for 20 min for the model to approach oil-wet conditions.
2. The model is carefully washed using methanol before being put in an oven to dry out.

Table 3
Seawater composition.

Salt	NaCl	KCl	$MgCl_2$. $6H_2O$	$CaCl_2$	Na_2SO_4	$NaHCO_3$
Concentration (ppm)	27 350	849	14 840	1330	4686	16.8

Table 4
Glass-micromodel tests properties.

Property	Value
Length	6 cm
Width	6 cm
Pore volume	0.5 cc
Porosity	39
Injection rate	0.85 cc/h
Pore volume injected	5

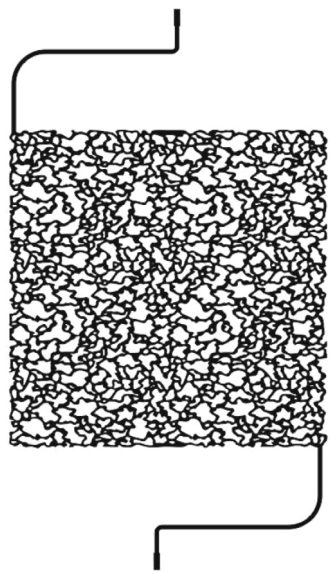

Fig. 1. A schematic of the heterogeneous pattern used in this study.

3. The model is saturated with the formation water.
4. The model is saturated with crude oil.
5. The model is flooded by the injection fluid (Seawater, Smart water and/or smart water-surfactant).

In order to evaluate the effect of the surfactants, smart water–surfactant samples with surfactant concentrations of 200, 500, 1000, 1500, 2000, 3000 and 5000 ppm were prepared and were used to flood the saturated glass micromodel. The recovery factors are calculated and recorded for each of the samples.

3. Results and discussion

3.1. Wettability

3.1.1. Effect of salts on wettability

To evaluate the effects of different salts present in the seawater on the wettability alteration capacity of it, smart water samples with changing salt concentrations were prepared and their wettability alteration were studied. In order to evaluate the effects of the five different salts, five different sets of samples were prepared. In each set, only one salt's concentration was changed, while the other salts' concentration remained the same as in the seawater composition. For instance, SW-2KCl is the sample in which the concentrations of all salts remain the same as in the seawater, while the concentration of KCl is twice as much as its concentration in the seawater. In each sample set, the concentration of the selected salt was fixed to zero, one, two, three and four times as much as its concentration in the seawater, while the other salts' concentration were fixed to theirs in the seawater. The results are displayed in Fig. 2.

As illustrated in Fig. 2, all salts display a peak in the contact angle implying that with increasing their concentration, the measured contact angle increases. After the peak contact angle, the contact angle decreases and hence the rock approaches water-wet conditions.

The results of this experiment are in great agreement with the literature. According to the literature, the surface of carbonate rocks are positively charged near the neutral pH value (Anderson, 1986). Given that SO_4^{2-} is a highly active anion, it can enter the double layer and bond with the positively charged Ca^{2+} cations on the rock surface, and hence freeing the oil's carboxylic groups formerly bonded with Ca^{2+}. Consequently, the oil molecules leave the rock surface and the water-wetness of the surface increases. This is why the more the concentration of SO_4^{2-}, the lower the contact angle of the rock.

This is also true for $CaCl_2$ and $MgCl_2$ since the rock becomes more water-wet as the concentrations of Ca^{2+} and Mg^{2+} increase. To explain this, the previously mentioned double layer theory can be used. As the sulfate ions continue to bond with the positively charged calcium ions on the rock surface, the rock surface's electrostatic repulsion force decreases and the positively charged bivalent cations can go further and more close to the rock surface. As a result of this, these ions can bond with some of the carboxylates formerly bound to the rock surface and hence cleaning the rock surface of the oil molecules. Given this, the double layer thickens and the rock approaches water-wetness (Fathi et al., 2011). The effects of both Ca^{2+} and Mg^{2+} are consistent with the results reported in the literature. However, the results obtained for KCl and NaCl were not in agreement with the literature. Although NaCl shows an adverse effect on the contact angle from 0aCl to 2NaCl, it starts to behave differently as the concentration increases more. In contrast to our results, other studies have shown that the lesser the NaCl and KCl concentrations, the more water-wet the rock becomes.

3.1.2. Effect of surfactants on wettability

Based on the previous measurements, the three most effective salts (the most contact angle alteration) in their optimum concentrations (the concentrations at which the least contact angles for each salt were obtained) were found to be 1NaCl, 3KCl and $4Na_2SO_4$. It is worth mentioning that the zero concentration for none of the salts were selected, as it is difficult to remove the salts present in the seawater for industrial purposes. Therefore, even though the measured contact angle for 0NaCl and $0CaCl_2$ is the lowest of all concentrations, these values cannot be selected as the optimum ones. In the case of NaCl, the lowest contact angle is achieved at 4NaCl. However, this cannot be selected as the optimum NaCl concentration because of the extremely high concentration of NaCl in the seawater. Given that the concentration of NaCl in the seawater is 27 350 ppm, a solution of 4NaCl contains 109 400 ppm NaCl, which is an extremely high concentration and it is not practical to prepare such solutions for the industrial use.

The effects of the surfactants used in this study (SDS and CTAB) on contact angle were studied using samples with different surfactant concentrations and the seawater with the optimum

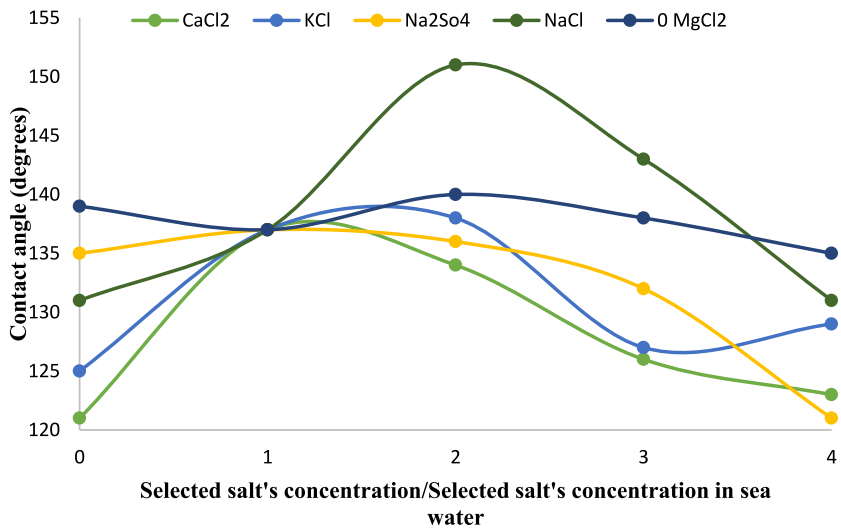

Fig. 2. The individual effect of salts on contact angle.

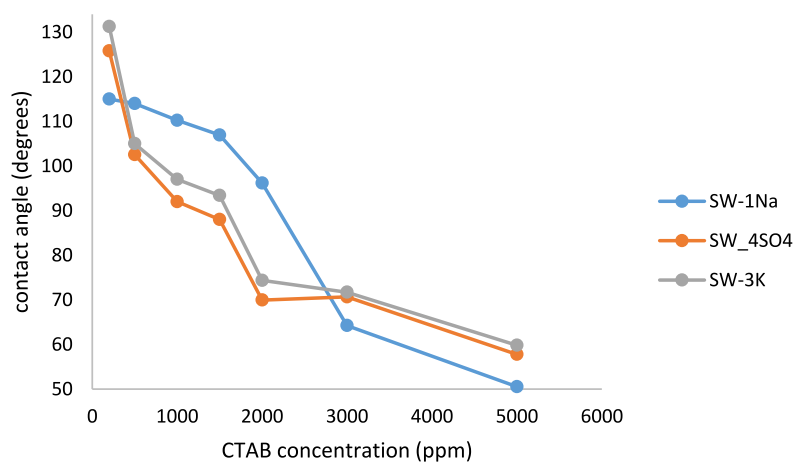

Fig. 3. The effect of CTAB on the measured contact angle in the modified smart water.

salt concentrations. Figs. 3 and 4 show the effect of CTAB and SDS on the contact angle, respectively.

As illustrated in Fig. 3, adding CTAB to the SW-1NaCl solution causes the contact angle to decrease and hence the rock to approach water-wet conditions. However, for the cases of SW-3KCl and SW-4Na_2SO_4, the contact angle does not demonstrate a monotonic decrease or increase with the CTAB concentration. In fact, not only the contact angle varies with the concentration, but also the behavior of the changes varies with it. At the critical micelle saturation of CTAB, SW-3KCl and SW-4SO_4^{2-} demonstrate a better performance, regarding their effects in lowering the contact angle of the brine. However, in extremely high concentrations of CTAB, SW-1NaCl has the most effect of the wettability of the rock. This shows that depending on the concentration of the salts in the seawater, the optimum concentration of CTAB changes.

As Fig. 4 demonstrates, increasing SDS concentration from 200 to 5000 PPM causes the contact angle to decrease. Nonetheless, the reductions in the measured contact angle do not behave monotonically for all the selected salts. For the case of SDS in the optimized samples of seawater, the behavior of contact angle changes. While at low SDS concentrations to CMC of SDS, SW-1NaCl shows the best performance in lowering the contact angles, at extremely high concentrations of SDS, SW-4SO_4^{2-} has the lowest contact angle. Overall, CTAB shows a better performance in lowering the contact angle of the rocks.

3.1.3. *Combined effect of CTAB and SDS on wettability*

Since the effect of surfactants on the wettability of the rocks is dependent on various parameters, such as the rock type, water salinity and temperature, any combination of surfactants and waters would have a different effect on the final recovery of the water flood (Mohammed and Babadagli, 2015). Therefore, in this study, we tried to find the best combination of the smart water and the surfactants with the highest wettability alteration potential for this study's specific rock.

In order to evaluate the combined effect of SDS and CTAB surfactants, a set of experiments were conducted. Table 5 shows the samples used in these experiments and their respective surfactant concentrations. The Critical Micelle Concentrations (CMC) for both CTAB and SDS are reported to be 364 ad 484 ppm, respectively. Moreover, the base solution for these measurements was the optimized smart water composition (1NaCl + 3KCl + 4Na_2SO_4).

As demonstrated in Fig. 5, mixing CTAB and SDS causes the overall contact angle of the prepared solutions to increase. In fact, a solution containing only 0.9CMC of CTAB and 0.1CMC of SDS is of the lowest contact angle. To illustrate more, 0.9 CMC of CTAB means that the solution has a concentration of CTAB equal to 0.9 of the CMC of CTAB. Therefore, in the case of this study, the best results are obtained by mixing 0.9CMC of CTAB and 0.1CMC of SDS in the optimized smart water. Given the results from these

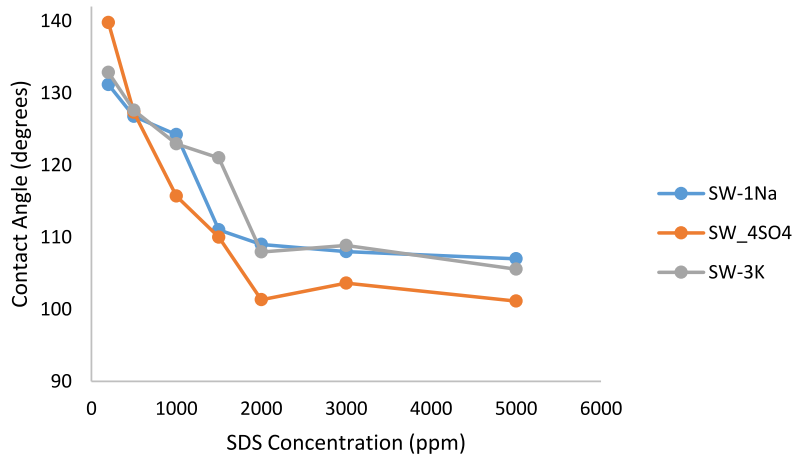

Fig. 4. The effect of SDS on the measured contact angle in the modified smart water.

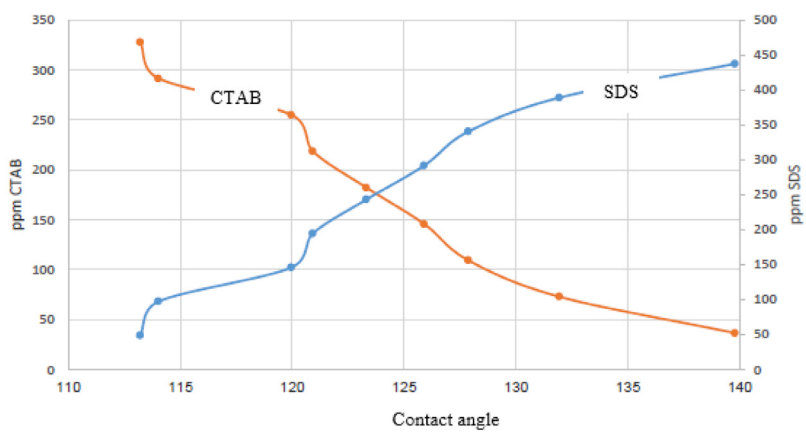

Fig. 5. Combined effect of SDS and CTAB on contact angles.

Table 5
The composition of the combinations of SDS and CTAB in optimum seawater.

CMC (SDS + CTAB)	Contact angle
48/6 ppm SDS + 327/6 ppm CTAB	113.2067
97/2 ppm SDS + 291/2 ppm CTAB	114.0217
145/8 ppm SDS + 254/8 ppm CTAB	119.9583
194/4 ppm SDS + 218/4 ppm CTAB	120.915
243 ppm SDS + 182 ppm CTAB	123.31
291/6 ppm SDS + 145/6 ppm CTAB	125.8917
340/2 ppm SDS + 109/2 ppm CTAB	127.865
388/8 ppm SDS + 72/8 ppm CTAB	131.94
437/4 ppm SDS + 36/4 ppm CTAB	139.7917

experiments, it is thought that for the case of this study (almost pure limestone), combining anionic and cationic surfactants has drawbacks. Nonetheless, since most reservoir rocks are a mixture of carbonate minerals, quartz and clays, there might be a possibility of mixing SDS and CTAB to achieve a higher overall wettability alteration.

3.2. Oil recovery factor

Using the aforementioned glass micromodel pattern, the recovery factor experiments were conducted.

To evaluate the effects of the salts and the surfactants on the ultimate oil recovery factor compared to the seawater's effect, first, an experiment was conducted to measure the recovery factor of the seawater. Distilled water flooding had a recovery factor of 28%. Changing the injection water to the seawater (Persian Gulf water) caused the oil recovery factor to increase to 42%. Then a combination of the optimum smart water and SDS were used as the injection water.

As can be understood from Fig. 6, increasing the concentration of SDS does not necessarily result in a better flood recovery. In fact, there is an optimum recovery factor for SDS which occurs at 500 ppm (which is close to CMC of SDS). Therefore, in the case of SDS and carbonate rocks, the optimum surfactant concentration is the critical micelle saturation.

The same sets of experiments were conducted for CTAB, too. Fig. 7 demonstrates the results of these experiments for CTAB. Similarly, the recovery factor for CTAB has an optimum value which occurs at 500 ppm. Overall, CTAB's recovery factor is higher than that of SDS.

4. Conclusion

Based on this research's results, combination of smart water and surfactants can be more advantageous compared to smart water flooding alone. As mentioned before, in the optimum surfactant and salt concentrations, the rock approaches water-wet condition, which can greatly assist the process of water flooding. Regarding the contact angle alteration capacity of the surfactants, CTAB has a greater potential to reduce the contact angle. Although not monotonically, the contact angle decreased with increasing the concentration of both surfactants when they were not used together in a sample. Water flooding with the optimum smart water has a higher recovery factor than seawater flooding. However, the highest recovery factor was attained using

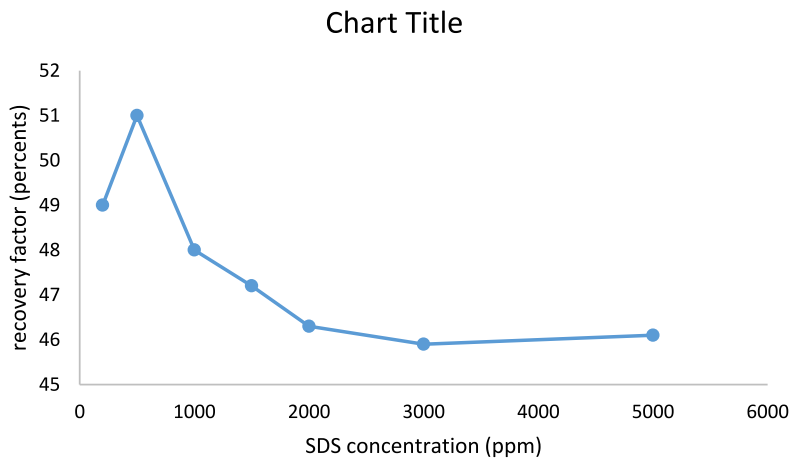

Fig. 6. Oil recovery versus SDS concentration.

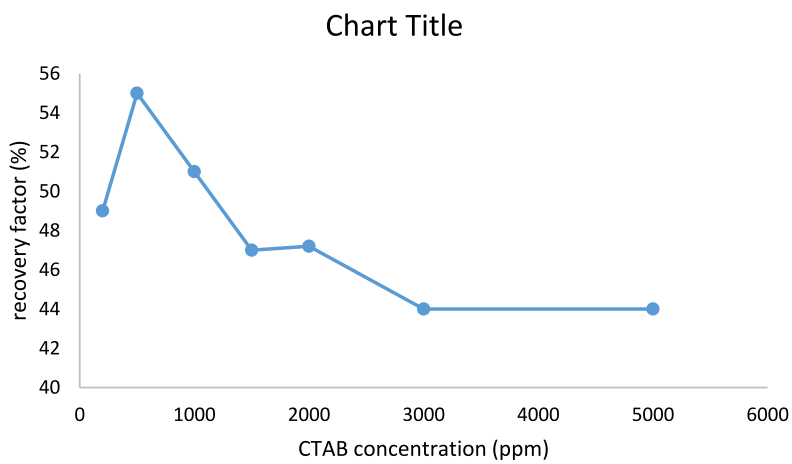

Fig. 7. Oil recovery versus CTAB concentration.

a combination of the optimum smart water and the optimum concentrations of the surfactants.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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