

## Review article

# A comprehensive review on interaction of nanoparticles with low salinity water and surfactant for enhanced oil recovery in sandstone and carbonate reservoirs



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## ABSTRACT

Nanoparticles (NPs) are currently gaining wide acceptance in the field of petroleum engineering. They are applied in different areas of petroleum exploration and production such as drilling, well logging, reservoir management, and enhanced oil recovery (EOR). Due to the size of NPs, they have special physical and chemical properties. Therefore, NPs can influence the properties of the fluid system, including viscosity, magnetism, and interfacial tension (IFT).

The injection of NPs into the reservoirs for EOR is more effective than water injection but not as effective as chemical flooding. Consequently, NPs are injected along with low salinity water (LSW) or chemicals such as surfactant in order to improve the recovery of oil. NPs are used to prevent the fines migration during LSW injection, control the mobility of formation water, and reduce the surfactant adsorption on the pore walls of the reservoir.

The improvement in oil recovery, when NPs are injected in combination with LSW or chemicals in the reservoir, can be attributed to the variations in the properties of the fluid system and the rock-fluid interactions. This study comprehensively reviews the mechanisms behind these variations. LSW injection improves the oil recovery by altering the rock wettability from oil-wet to water-wet. However, this study reveals that the dispersion of NPs in LSW does not necessarily change the rock wettability towards water-wet. The wettability of the system may shift towards oil-wet instead of water-wet depending on the concentration of NPs. Improvement in oil recovery depends on the effective surface charge and the volume fraction of dispersed NPs in the solution. Aggregation of NPs in solution should be avoided because it lowers the recovery by plugging the pore throats. The stability of NPs dispersed in solution with increasing concentrations of salt and surfactant is reviewed and the resulting effect on the IFT of the solution is analyzed. NPs dispersed in different types of surfactant show different behaviors of IFT. The behavior of the IFT depends on the concentration of surfactant, the amount of dispersed NPs (concentration), the type of surfactant (anionic, cationic, and non-ionic), and the effective charge of NPs (positive, negative, and neutral). The combination of LSW with surfactant for oil recovery has two opposing impacts. The IFT reduces while the contact angle increases with an increase in salinity.

The mechanisms responsible for the variations in the properties of the system when the combination of LSW, surfactant, and NPs are used for oil recovery are reviewed in this study. Understanding the mechanisms behind the interactions at the fluid-fluid and the fluid-solid interfaces will aid in designing an effective fluid system that combines LSW, surfactant, and NPs for successful implementation of EOR in sandstone and carbonate reservoirs.

## 1. Introduction

Due to the size of Nanoparticles (NPs), they exhibit special physical and chemical properties [1]. The NPs size varies in the range of 1–100 nm, which enhances their application in numerous professional fields. They have gained wide acceptance in different fields of science

and engineering like pharmacy, medicines, ceramics, and metallurgy. The application of NPs to hydrocarbon reservoir formations has increased because of their mechanical and thermal stability properties [2,3].

The advancement in technology has made the manufacturing of NPs to be easy and cost-effective [4]. This increases the application of NPs

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to the oil reservoirs for enhanced oil recovery (EOR) [5]. Several studies have shown that a dispersed solution of NPs is efficient to alter the wettability of the rocks towards water-wet [6–9] and reduce the interfacial tension (IFT) [2,10]. However, NPs become more popular in the petroleum field as a result of their capacity to prevent fines migration in the reservoir during low salinity water (LSW) injection [11–18]. Currently, NPs are being combined with LSW or chemicals used in EOR such as surfactant and alkaline to alter the rheological properties of the fluid system like viscosity, control and reduce the surfactant adsorption on the pore walls of the reservoir and the fines migration, and enhance oil recovery.

During the past three decades, surfactant flooding has been implemented for tertiary recovery of oil in the depleted reservoirs. Surfactants reduce the IFT between brine and oil; thus, enhance recovery and lower the residual oil saturation [19,20]. However, the surfactants are significantly absorbed by the porous media, which affects their capability to reduce the IFT [21]. Several studies have also revealed that there are optimum water salinity and optimum temperature for effective performance of surfactants. The optimum water salinity for effective performance of surfactants is lower than the reservoir brine salinity [22–24]. This impacts the effectiveness of surfactants for EOR at the field scale.

In order to improve the effectiveness of surfactants for tertiary oil recovery, various studies were conducted to show that the adsorption of surfactants on the walls of the pores is reduced by addition of NPs to surfactants [25–28]. Zargartalebi et al. [10] observed that the adsorption of surfactants and the IFT between brine and oil can be drastically reduced with addition of NPs at a concentration below the critical micelle concentration (CMC) of the surfactants.

LSW injection into the reservoir formations has been a subject of discussion within the past two decades. The laboratory and the field implementations of the LSW injection into sandstone and carbonate reservoirs have been reported to be successful [29–33]. Also, different theories and hypotheses have been proposed as the drive mechanisms for tertiary recovery of oil during LSW injection. Some of the popular proposed hypotheses are the reduction of the IFT, the multi-ion exchange, the electrical double layer (EDL) effect, the salt-in effect, and the osmotic pressure [34,35]. Fines migration has been argued to be vital in the recovery of oil especially in sandstone formations [32]. However, the results of some conducted experiments are in contradiction with the theory that fines migration contributes to more oil recovery during LSW injection [36–40]. Fines migration occurs due to a decrease in the brine salinity and an increase in the pH. Despite the arguments on the mechanisms used by LSW to improve oil recovery, there is still a consensus that LSW flooding alters the wettability of the rocks towards water-wet [38,41–43].

NPs are currently being used in the treatment of fines migration during LSW injection [12,15,18,44,45]. However, the selection of the appropriate NPs to reduce fines migration in the reservoir is essential to the success of LSW injection. The desired NPs prevent the fines from damaging the formation by plugging the pore throats. Several approaches for the treatment of fines migration by NPs during LSW injection were proposed in order to assess the efficiency of the NPs [38,41–44].

Also, combination of LSW with surfactant injection has been applied to both carbonate [46,47] and sandstone [48] formations. The injection of surfactant improves the oil recovery from the reservoirs by reducing the IFT between oil and water. The combination of LSW with surfactant injection improves oil recovery by altering the wettability of the rock and reducing the IFT of the fluid system [46–48].

Although the injection of NPs in combination with LSW or chemicals (such as surfactant) has been reported to be successful for oil recovery from different reservoir formations, there is no consensus on the most suitable NPs for a particular reservoir and the best scenario to apply NPs injection. Fig. 1 explains the different combinations of LSW, surfactant, and NPs that enhance the oil recovery from different

reservoir formations. The combination of two or three solutions of LSW, surfactant, and NPs improves the performance of the fluid system. However, the best combination and the most effective injection pattern for the combination of LSW, surfactant, and NPs for EOR are still vague. Understanding the mechanisms behind the interactions at the fluid-fluid and the fluid-solid interfaces is a guide for proper designing of an EOR method.

This work comprehensively reviews the mechanisms underlying the application of NPs in combination with LSW or chemicals like surfactant to the oil reservoirs. The review is divided into different parts in order to explain the mechanisms behind all the possible combinations of LSW, surfactant, and NPs used in the EOR techniques.

First, the stability of NPs in low and high salinity water is discussed to understand the behavior of the interaction between NPs and salt particles. Several techniques have been devised to determine the stability of NPs in solution. However, this work tries to relate the effects of the concentration of ions in solution and the concentration of NPs on the stability of NPs in solution. The significance of the zeta potential for determination of the NPs efficiency in reduction of fines migration is discussed. The variations in the contact angle and the oil recovery factor when a solution of NPs is injected into the reservoir is also reviewed using the results of the experiments reported by several studies in literature.

Then, the interaction forces that play critical roles in the recovery of oil when NPs is injected along with surfactant into the reservoir is discussed. Several results from existing experimental studies have shown that both the surface charge of selected NPs and the type of surfactant affect the IFT. The mechanisms responsible for the variation in the IFT are discussed.

Finally, the contributing mechanisms when LSW is injected along with surfactant to modify the wettability of the rock and reduce the oil-water IFT are reviewed. The basic screening criteria for the injection of NPs with LSW or surfactant into the reservoirs is thoroughly discussed and highlighted in this study.

This article is organized as follows. First, the interactions of NPs with deionized water and low and high salinity water are reviewed. Later, the interaction of NPs with surfactant is studied. Then, the interaction of LSW with surfactant is discussed. Finally, the summary and conclusions are presented.

## 2. NPs dispersed in deionized water and low and high salinity water

The Derjaguin-Landau-Verwey-Overbeek (DLVO) and the non-DLVO theories play important roles in the stability of NPs [49]. The DLVO theory states that the balance between the attractive forces (dispersion forces) and the electrostatic repulsive forces enhances the colloidal stability. An energy barrier created by the repulsive forces, which prevent the approaching of two particles to each other, results in a stable colloidal solution. Meanwhile, the attractive forces bring the particles together to overcome this barrier, which results in the coagulation of particles. The attractive forces (Van der Waals forces) are due to the electrical and the magnetic polarization properties of the ions of particles.

The stability of NPs in solution of deionized water and brine is attributed to these forces. The stability of NPs increases with an increase in the surface charge of NPs (zeta potential) and a decrease in the salinity of the water [50]. Aside the DLVO forces, other forces, which refer to the non-DLVO forces, also affect the stability of the solution. The non-DLVO forces are the steric forces, the magnetic forces, and the hydration forces [49]. Steric forces occur when the particles adsorb some organic matters like surfactant to stabilize the solution. The magnetic forces are due to the ability of the particles to create a magnetic dipole moment in the absence of applied magnetic field for stabilizing the solution (for example iron, nickel, cobalt, and manganese). Zeta potential is used as a measure of balance between the DLVO forces

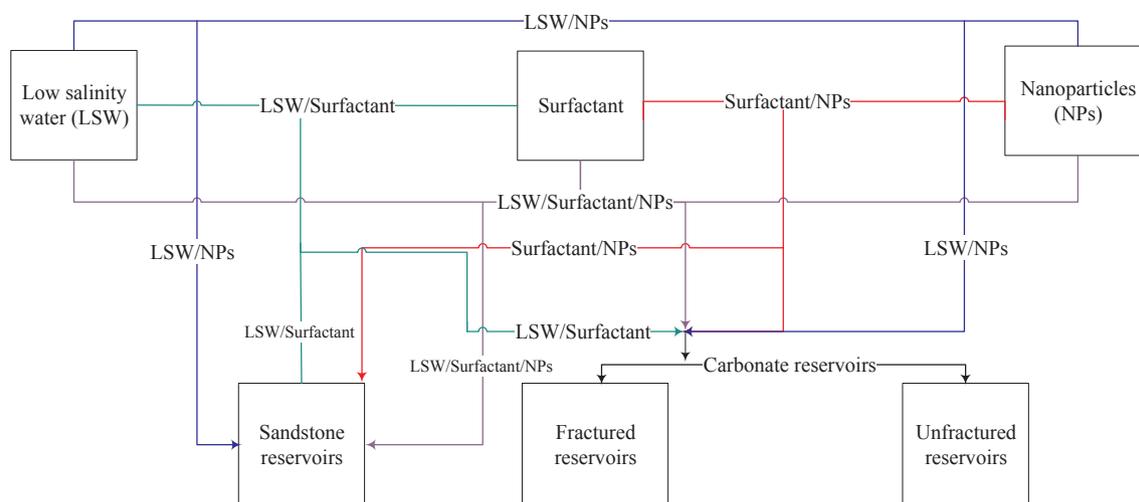


Fig. 1. Enhanced oil recovery from sandstone and carbonate reservoirs using different combinations of LSW, surfactant, and NPs.

and the non-DLVO forces that enhances the stability, the surface potential, and the surface charges. The detailed formulations for the DLVO and non-DLVO forces are presented by Petosa et al. [49].

### 2.1. Behavior of NPs in deionized water

The colloidal solution of NPs is stable in deionized water at room temperature. The NPs hydrolyzed in water form a variety of compounds. For example, silica ( $\text{SiO}_2$ ) hydrolyzed in water form silanol and silane groups depending on the temperature. The amount of silanol groups in the solution affects the pH, the ionic strength, the surface potential, and the wettability alteration strength of the NPs. Grabbe and Horn [51] explained that a repulsive force acting at a short range distance, which is different from the electrostatic forces of repulsion, is discovered in the solution of silica NPs [52]. The solution is dominated by the repulsive forces and the Van der Waals forces of attraction. The repulsive forces can be referred to the hydration forces (non-DLVO forces) of the particles. Therefore, the composition of the ions present in the dispersed solution of NPs is vital to the stability of the solution. Some cations like Si and Zn have the ability to form a complex compound while there are other cations that form compounds with specific properties or surface charges. For example, alumina ( $\text{Al}_2\text{O}_3$ ) hydrolyzed in water reaches an equilibrium with  $\text{Al}(\text{OH})_3$  [53]. The charges present in deionized water are  $\text{OH}^-$  and  $\text{H}^+$  ions, which are attached to the water molecules by hydrogen bond. Dispersion of NPs in water distorts the orientation of the ions and as a result both the  $\text{OH}^-$  and  $\text{H}^+$  ions move toward the particles depending on the surface charge. The charge on the particle surface results in either repulsion or attraction of the  $\text{OH}^-$  or  $\text{H}^+$  ions [52,54]. Zargartalebi et al. [55] observed that two different solutions of ninety percent pure silica NPs have different colloidal stabilities in water. The colloidal stability of the silica NPs in deionized water is due to the effective charges on the silica NPs. The effective charges on the silica NPs allow the repulsive forces to overcome the attractive forces, which enables the NPs to attain a long stability in solution [56]. Therefore, the surface charges of dispersed alumina NPs in deionized water are positive because the zeta potential measured is positive while the surface charges of dispersed silica NPs in deionized water are negative because the zeta potential measured is negative [57]. For example, silica NPs are slightly adsorbed by limestone formation while they are not retained in quartz and sandstone formations [49].

### 2.2. Behavior of NPs in low and high salinity water

In the presence of other ions, the mechanisms involved in the

stability of the colloidal solution of NPs become more complicated. This results in either stabilization or coagulation of the NPs in solution. Unlike the deionized water, there is an exchange of varieties of charges and ions in the solution, which depends on the activity (concentration) of the particles and the pH of the colloidal solution [54]. The salt ions prevent the repulsion of particles by screening them, which leads to coagulation of the NPs [56]. For a solution of low salinity, the system is dominated by proton exchange, which increases the surface potential. However, the activity of the adsorbed ions reduces with increasing the surface potential and the dissociation group [58]. It has been reported that the pH of colloidal solution reduces with an increase in the concentration of the NPs [59,60]. The colloidal stability of the silica NPs in sea water can be improved by adding acid to the solution. The  $\text{H}^+$  ions of the acid protect the silica NPs from the ions of the sea water [61]. It is noted that the pH depends on the amount of cations and NPs ions in the solution at a specific temperature. Skauge et al. [62] reported that 300 ppm silica NPs is stable in 0.5 wt% of NaCl while Yousefvand and Jafari [7] explained that aggregation of NPs occurs when 3 wt% of NaCl is added to 0.5 wt% of silica NPs. The salt acts as the controlling parameter, which causes the NPs to aggregate 0.07 wt% of the NPs. This behavior is attributed to the screening of charges and the EDL due to the high salinity concentration [7]. Abbas et al. [56] explained that the presence of salt ions increases the viscosity and the refractive index of NPs dispersed in solution. The aggregation of NPs is attributed to an increase in the attractive forces different from Van der Waals forces due to the screening effect between the particles and the salt ions [56]. Esfandyari et al. [57] reported that the stability of NPs dispersed in saline solution has a direct relationship with the zeta potential. Solutions of dispersed NPs are stable in deionized water when the zeta potential is extremely positive or negative. The zeta potential is directly related to the surface charge of the NPs in solution. Addition of salt to the solution alters the value of the zeta potential towards zero where aggregation of NPs occurs in the solution [63]. The size of the NPs is also an important factor, which affects the stability of the colloids. The colloidal solution is more stable for smaller sizes of NPs depending on the activity of the salt [63].

### 2.3. Retention of fines migration by NPs

It has been widely reported that NPs can reduce fines migration which occurs during LSW injection into the reservoir [64,65]. This is a frequent problem in sandstone formation due to the presence of clay and feldspar. Numerous studies have been conducted to determine the most suitable NPs that can adsorb fines in a sandstone formation. Assef et al. [16] reported that the valence of the ions present in the solution

influences the adsorption of fines by NPs. The interactions of the NPs with the rock surface and the aqueous solution have been investigated using several techniques like atomic force microscopy (AFM), quartz crystal microbalance (QCM), and X-ray diffraction (XRD) to determine the key controlling parameters in fines migration and wettability alteration.

In a reservoir system, the geochemical properties of the rock, the polar components of the crude oil, and the ions of the formation brine present in the reservoir are in chemical equilibrium, which prevents the dissolution and the precipitation of the chemical components [32]. The chemical equilibration of ions is distorted by the injection of LSW into the reservoir. This leads to the dissolution and the precipitation of chemical compounds, which are transported along with the fluids. The transportation of the dissolved chemical components is referred to fines migration. Several studies have reported that fines migration is only observed in formations containing clay [44] when LSW is injected into the reservoir. The types of minerals (clay and feldspar) present also affect the formation of fine particles. Barnaji et al. [44] explained that the non-swelling clay (kaolinite) enhances the formation of fine particles that increases the oil recovery while the swelling clay (sodium bentonite) reduces the permeability by plugging the pore throats.

The processes involved in the formation of fines migration when LSW is injected into the reservoir are significant to understand the mechanisms behind the reduction of fines migration by NPs dispersed in solution. Injection of LSW into the reservoir changes the distribution of the ions present in the reservoir brine [66]. The change in the distribution of ions causes protonation or deprotonation of  $H^+$  ions; therefore, the pH of the effluent changes [61]. Clay and feldspar are composed of aluminum and silicate with a trace amount of iron, alkali (Na, K), and alkali earth metals (Ca, Sr, Mg), which are dissolved because of a change in the ionic strength of the solution due to LSW injection. In a reservoir containing clay, the brine forms an equilibrium with the system, which makes the surface charges of the system positively or negatively charged depending on the electrical balance of the ions in the system. The equilibrium system prevents the continuous dissolution and precipitation of the clay in the system. Injection of the LSW changes the distribution of ions in the equilibrium system, which causes the dissolution of the clay or feldspar particles. This occurs at a concentration smaller than the critical salt concentration of the monovalent ions ( $Na^+$ ) [66]. The dissolved clay particles are attracted to the cations and anions present in the solution to form insoluble compounds that are transported as fines. For example, Barnaji et al. [44] explained that the crude oil containing carboxylic components attaches to the clay surface when the reservoir rock has no connate water. In this case, the crude oil is detached from the clay surface during the injection of LSW. The crude oil does not attach to the clay surface when the reservoir rock is initially flushed with brine (or when the reservoir rock has connate water). Fine particles are produced in both cases, but improvement in oil recovery is only observed in the reservoir rock containing clay without connate water [44].

It is inferred that the clay surface is positively charged in the absence of connate water [44] while the carboxylic group present in crude oil is negatively charged; therefore, the crude oil is attracted to the clay surface. It is essential to note that the brine and the crude oil components are not the only factors which cause the clay surface to attract the crude oil. Injection of LSW into the reservoir dissolves some chemical components of the clay that leads to the attraction and the repulsion of different ions. The clay surface is changed from positively charged to negatively charged or neutral to release the crude oil when LSW is injected. Consequently, fines migration occurs when the interaction forces between the fluid and the particles are repulsive, which leads to coalescence of the energy barrier.

It can be concluded that all the mechanisms above-mentioned are responsible for oil recovery when LSW is injected into the reservoir. It has been reported by several studies that different types of NPs can be efficient in controlling fines migration [12,15,16,18,44,45]. The NPs

are attracted to the surface of the pore walls. This subsection explains the mechanisms involved to reduce fines migration by alumina and silica NPs.

Alumina NPs dispersed in deionized water have positive charges that dominate the colloidal solution while silica NPs dissolved in deionized water have negative charges that dominate the colloidal solution. The surface charge of NPs varies with an increase in salinity [16,52]. The NPs affect the ionic strength of the solution, which results in controlling the fines migration. Increasing the ionic strength of solution by adding  $\gamma\text{-Al}_2\text{O}_3$ , silica, and CuO NPs in LSW increases the adsorption of fine particles while the ZnO and MgO NPs dispersed in deionized water and LSW have a slight effect on the adsorption of fine particles [15]. The retention of fines by the solutions of  $\gamma\text{-Al}_2\text{O}_3$ , silica, and CuO NPs is attributed to a decrease in the size of the NPs when dispersed in LSW compared to when dispersed in deionized water. However, the size of ZnO and MgO NPs increases when dispersed in LSW compared to when dispersed in deionized water. The reported size of the NPs in both deionized water and LSW is larger than the reported size by manufacturer ( $< 100$  nm). The increase in the size of the dispersed NPs is attributed to the attraction of the ions with different surface charges to balance the ionic repulsive forces that enhance NPs stability in solutions. For example, Arab et al. [15] showed that fine particles, containing different oxides of metals and non-metals ( $> 80\%$   $\text{SiO}_2$ ), dispersed in deionized water and LSW have a negative zeta potential. However, the fine particles dispersed in the LSW are more positive than the fine particles dispersed in the deionized water. Also, the zeta potential of silica beads used during the experiment is more positive ( $-27.6$  mV) in LSW compared to the one in deionized water ( $-44$  mV). The amount of particles trapped in the porous media when the solution has no NPs is dominated by the advective and diffusive forces [67], which only adsorb a small amount of the fine particles [15].

The surface charge of alumina NPs (positive) dispersed in solution overcomes the energy barrier existing between the surface of the pore walls and the clay in the reservoir. Therefore, the alumina NPs are adsorbed at the surface of the pore walls. As a result, the negative surface charges [68] are changed to the positive surface charges. This causes the attraction of the fine particles to the surface of the alumina NPs. For example, immersion of silica beads in solution of alumina NPs dispersed in LSW has more positive surface charges compared to immersion of silica beads in deionized water [15]. Consequently, most of the particles are retained in the porous media. It is inferred that the ability of NPs to control fines migration depends on the resultant surface charges of the NPs, the surface charge of fine particles in ionic solution, and the fluid flow velocity. For example, a solution of alumina NPs with the highest positive zeta potential has the most retentive capacity of fine particles in silica beads while a solution of ZnO NPs with the most negative zeta potential has the least retention of fines in silica beads as shown in Figs. 2, 3(a), and (b). The most important factors to control the fines migration in the reservoir are the resultant electric charges of the ions in the solution and the types of fine particles that are formed in the ionic solution. For example, Assef et al. [16] demonstrated that the surface charges of MgO NPs, which were reported not to be effective for reduction of fines migration [15], can be improved by increasing the ionic strength of MgO NPs dispersed in solution. Fig. 3(a) and (b) illustrate the capacity of MgO NPs to retain the fine particles with an increase in the ionic strength ratio of MgO NPs dispersed in solution. The ionic strength ratio of NPs is expressed as the ratio of the NPs ionic concentration dispersed in solution to the total ionic concentration in the solution. The MgO NPs behave differently in solutions containing monovalent and divalent metal ions of chloride with the same concentration. The capacity of MgO NPs to adsorb fine particles is highest when the zeta potential of the solution is the most positive as shown in Fig. 4(a) and (b). Hence, the adsorption of fine particles by NPs in the porous media depends on the concentration of NPs dispersed in solution and the effective surface charges, which balance the repulsive and attractive forces between the NPs and the fine particles

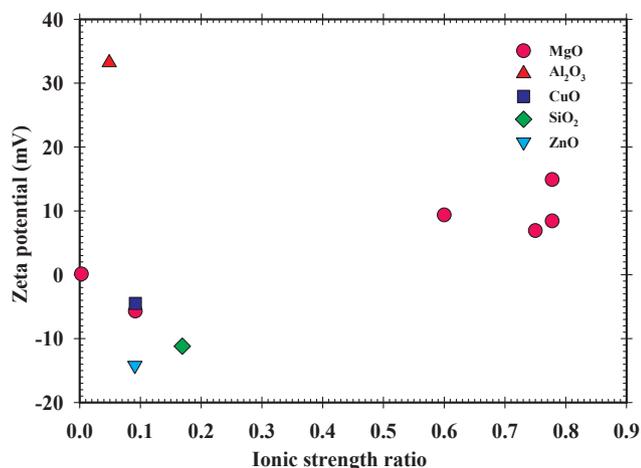


Fig. 2. The zeta potential of the silica beads in different solutions of NPs versus the ionic strength ratio of NPs to control fines migration. The experimental data used here are extracted from the works conducted by Arab et al. [15] and Assef et al. [16].

[16].

#### 2.4. Wettability alteration by NPs

Wettability alteration of the reservoir rock is affected by the brine composition, the polar components of the crude oil, the surface roughness, the temperature of the reservoir, and the minerals in the rock. The interactions of the minerals in the rock with the brine and the polar oil molecules are measured in terms of the zeta potential [69].

Alteration of the wettability by NPs is attributed to the adsorption of NPs by the reservoir rock [6]. The adsorption of NPs to the rock surface is because of the surface forces, which are the colloidal and hydrodynamic forces [45]. The surface charge of a reservoir rock is a function of the ions present in the brine, the temperature, the pressure, and the composition of the minerals in the reservoir rock. NPs have a large specific surface area, which aids their adsorption to the rock surface. NPs alter the wettability of reservoir rock by changing the surface charge on the pore walls, which can result in improving the recovery of oil. Li and Cathles [70] showed that the surface charge of a carbonate rock can be changed from positive to neutral by increasing the SO<sub>4</sub><sup>2-</sup> of the brine composition. Thus, the wettability alteration of reservoir rock by NPs is described using the DLVO and non-DLVO theories.

In a natural reservoir, the surface charges of the minerals in the rock are balanced by the brine ions and the polar oil charges in the EDL. Injection of NPs into the reservoir to alter the rock wettability exhibits both the electrophoresis and the streaming potential during the process, which are used to measure the zeta potential [69]. The electrophoresis is relative movement of the particles/charges to the stationary reservoir while the streaming potential is the normalized potential difference between the moving particles/charges and the stationary reservoir [69]. The surface charges of the reservoir rock depend on the relative concentrations of H<sup>+</sup> and OH<sup>-</sup> ions in the solution, the mineral compositions of the system, and the temperature. For example, the surface charge of calcite formation is independent of pH but depends on the concentration of Ca<sup>2+</sup> and CO<sub>3</sub><sup>2-</sup> in the equilibrium solution [69]. The attraction of NPs to the pore walls is an interaction between the NPs and the rock surface, where the energy barrier separating the fluid-rock interface is overcome, which results in wettability alteration [49]. The adhesion of fluid to the NPs coating the pore walls enhances the wettability alteration of the pore surface [71].

Increasing the concentration of NPs in solution does not necessarily enhance the wettability of the rock towards water-wet. The alteration in the wettability depends on the force balance in the triple line among the solid, the brine, and the NPs in the system [71]. The acidic groups present in the crude oil are very important in order to choose the suitable NPs for wettability alteration. Several studies have reported an optimum concentration of NPs in different solutions for the alteration of wettability using various techniques. The methods used to determine the optimum concentration of NPs are the spontaneous imbibition [72], the oil recovery factor from core flooding [73], and the contact angle measurement [72,74]. Fig. 5 demonstrates the variation in the contact angle of the system with an increase in the concentration of the NPs. The contact angles reported by different studies vary between extreme values with increasing the concentration of the NPs. The coalescence and the aggregation of NPs at a high concentration of NPs, which result in the plugging of pore throats, the fines migration, and the permeability reduction, are responsible for the cyclic behavior in the contact angle and the oil recovery factor highlighted in Fig. 5 and 6, respectively [72,74]. Despite an increase in the concentration of NPs, Youssif et al. [73] attributed the reduction in oil recovery (which happens after the maximum value of the oil recovery) to fast coagulation of the NPs, which plugs the pore throats [75].

The particle-fluid interactions between the ions of NPs and brine, the size of NPs, and the surface roughness of particle-particle interactions affect the wettability alteration [74]. Wettability alteration occurs when the repulsive forces between the rock surface and the NPs and

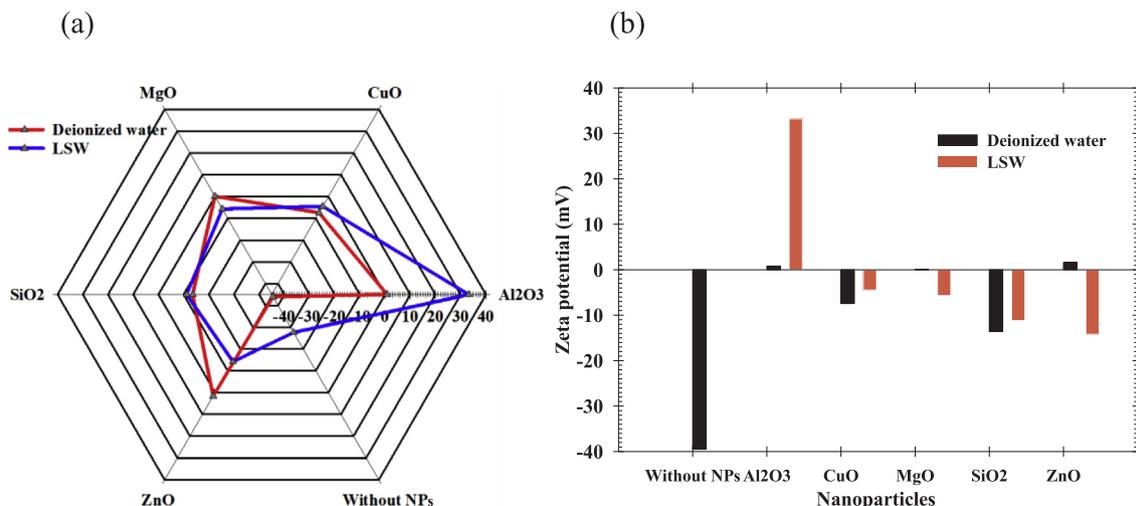


Fig. 3. The radar chart (a) and the bar chart (b) show the zeta potential of different NPs dispersed in deionized water and LSW (NaCl solution) to reduce fines migration in silica beads. The experimental data used here are extracted from the work conducted by Arab et al. [15].

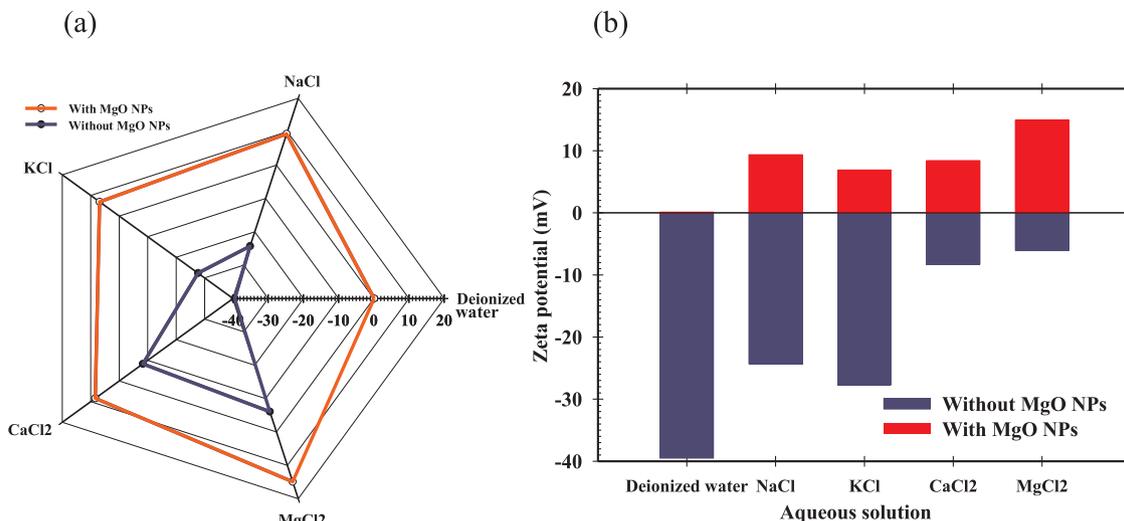


Fig. 4. The radar chart (a) and the bar chart (b) exhibit the zeta potential of the silica beads in different salt solutions with and without MgO NPs to attract fine particles. The experimental data used here are extracted from the work conducted by Assef et al. [16].

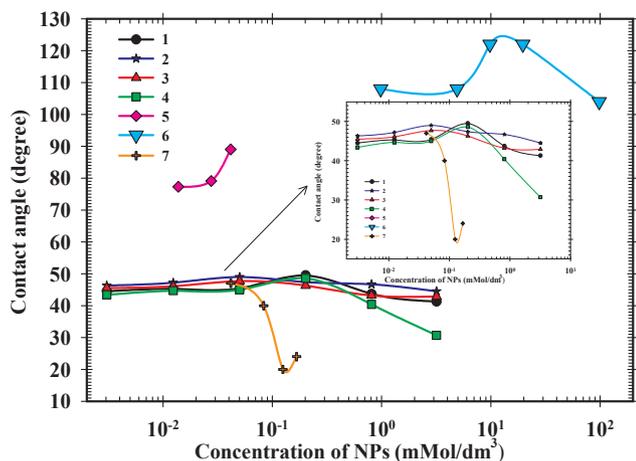


Fig. 5. The variation of the contact angle versus the concentration of NPs. The legends (1–4), (5 and 6), and 7 are the experimental data reported by Kim and Kim [71], Giraldo et al. [72], and Yousefvand and Jafari [7], respectively.

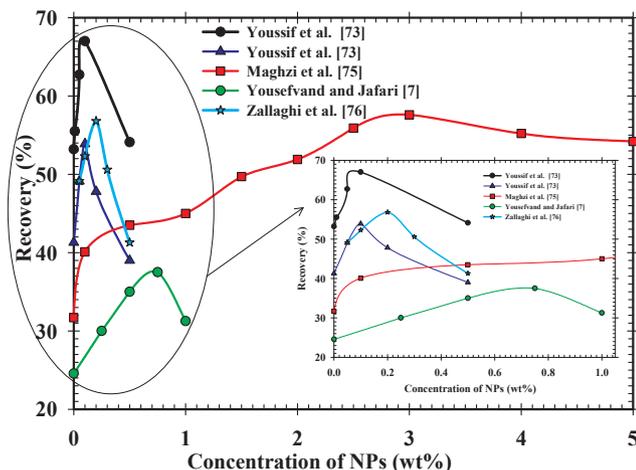


Fig. 6. The recovery factor versus the concentration of NPs. The experimental data were reported by different studies [7,73,75,76].

attractive forces between the NPs and the brine are balanced [74]. Consequently, the disjoining pressure and the contact force of the particle-particle and particle-fluid interactions, which prevent coalescence of the droplets from forming an interdroplet layer, diverge the wettability of the medium [77,78]. The thin film of the droplet between the particle interfaces is formed when the contact force (compressive pressure) and the disjoining pressure are balanced [79]. The volume fraction (concentration) of NPs dispersed in solutions has a relationship with the thermodynamic properties, which are affected by the disjoining pressure and the contact force [79]. Princen [79] showed that the contact angle has a direct relationship with the volume fraction of the particles. The volume fraction of the particles acting on the thin film depends on the contact force in the colloidal system. There is a maximum disjoining pressure beyond which the film collapse refers to the film rupture [79]. The film rupture leads to change in the droplet radius, which has an inverse relationship with the contact force. In addition, the thickness of the film is also a function of the droplet radius and the volume fraction of the particles. However, the thickness of the film does not depend on the contact force.

The oscillatory force of the particles is an important force that also affects the wettability of the rock. The oscillatory force occurs when the surfaces of two particles are moved close to each other [80]. The average periodicity of the force is related to the mean diameter of the molecule and the bound rigidity with the surface [80]. The oscillatory force decays with an increase in the separation of the particles to reach the Van der Waal force. The Van der Waals force is the only force occurring in a liquid where the particles have no ordered structure. Kralchevsky and Denkov [81] showed that the oscillatory force is dependent on the film thickness of the droplet and the particle volume fraction (concentration). The disjoining pressure is related to the oscillatory force as follows [81,82]:

$$\Pi = \Pi_{vw}(h) + \Pi_{el}(h) + \Pi_{os}(h) \tag{1}$$

where  $\Pi$  is the disjoining pressure,  $h$  is the film thickness of the droplet,  $\Pi_{vw}$  is the van der Waal force,  $\Pi_{el}$  is the electrostatic repulsive force, and  $\Pi_{os}$  is the oscillatory force which is zero for liquid particles without an ordered structure.

NPs spread across the edge of the film droplet in contact with the three-phase contact region in order to form a wedge as shown in Fig. 7 [77,83]. The particles in the wedge have the highest pressure in the vertex (plateau region) with oscillatory variation in the disjoining pressure due to an increase in the centre-to-centre distance between particles [77]. Therefore, the free energy of the system also varies in the

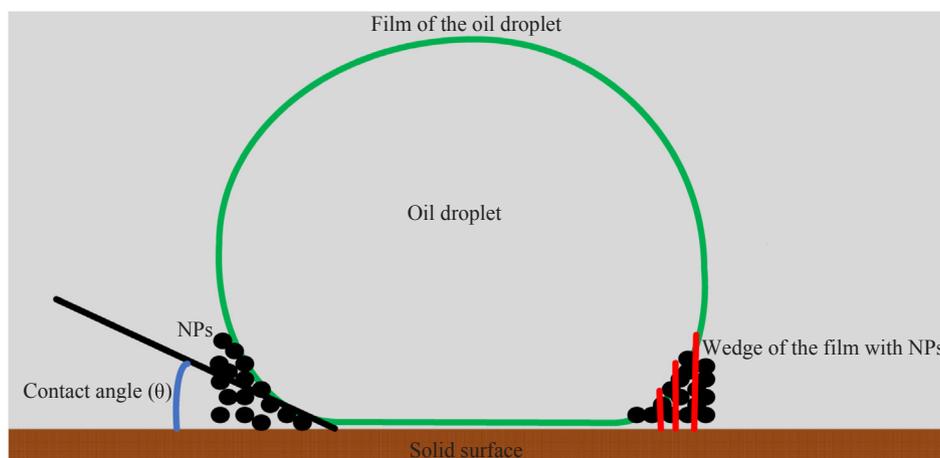


Fig. 7. The ordering structure of NPs in the film wedge that causes variation in the contact angle of the film surface.

oscillatory pattern, which results in several minimal and maximal values. The reduction of the contact angle at the interface with time for a solution of NPs reported by Al-Anssari et al. [84] can be related to the spread of the particles on the wedge by different forces (including viscous, capillary, diffusive, and gravitational forces) [85], which cause reorientation of the structure of the NPs [86]. The spreading coefficient can be related to the surface tension (interfacial tension) of the film and the concentration of NPs in the solution.

Consequently, the variation in contact angle with an increase in the concentration of the NPs is attributed to the oscillatory force of the disjoining pressure existing in the particle, the volume fraction (concentration) of the NPs, and the thickness of the film. The reduction in contact angle (including static equilibrium, advancing, and receding) with time is due to the gravitational force, which causes re-ordering in the structure of NPs.

### 3. NPs and surfactants

Surfactants are combined with NPs when an experiment is conducted in the laboratory for EOR [84]. However, the main objective of adding surfactants to NPs is to ensure the stability of NPs, which are unstable at a high concentration of salt solutions [87]. Recently, several studies have investigated the behavior of the adsorption of surfactant-NPs solutions in the porous media and the variation in IFT [76]. The mechanisms involved in the stability of NPs dispersed in a solution of high salinity and the variation of IFT with different surfactants are highlighted in the following subsections.

#### 3.1. Stability of NPs with surfactants

NPs have a high surface area to volume ratio and a high surface energy that make them to aggregate in high concentration of salt solutions [88]. Surfactants are added to a dispersed solution of NPs to stabilize the solution. In addition to stabilization, surfactants also modify the surface charges of NPs [88]. As above-mentioned, several forces affect the stability of NPs in aqueous solutions. Addition of surfactants to the dispersed solutions of NPs introduce the non-DLVO forces, which refer to steric forces [89].

A dispersed solution of NPs is stabilized through the adsorption of surfactant to the particles to form monolayers [90]. The amount of surfactant adsorbed increases with an increase in the concentration of surfactant until the critical micelle concentration (CMC) is reached [91]. Adsorption of surfactant on the surface of the NPs enhances the hydrophilicity of NPs [91]. The amount of surfactant adsorbed by the molecules depends on the hydrophilic-lipophilic difference (HLD) [87] and the radical group, like ethoxylated oxide (EO), of the surfactant

[92], the degree of NPs hydrophobicity [92], and the concentration of the brine [93]. The stabilization of NPs in the aqueous solutions is determined by the amount (thickness) of surfactant adsorbed and the type of surfactant [91].

The resultant surface charges of the dispersed solution of NPs is important in selecting the appropriate surfactant type for the stabilization. For example, Binks et al. [90] showed that addition of a cationic surfactant to a solution of silica NPs (negatively charged) causes sedimentation of the NPs in the solution. The zeta potential shows that the addition of surfactant and solution of NPs, which are oppositely charged, reverse the surface charges of the solution [27,90].

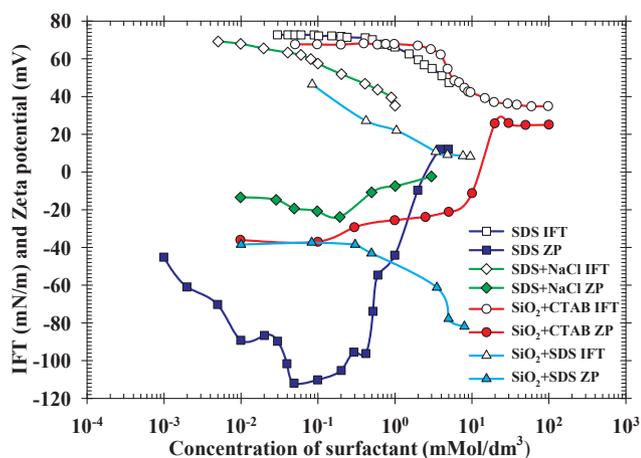
#### 3.2. Variation in IFT for solutions of surfactants and NPs

The addition of NPs to a surfactant solution has impacted the IFT behavior of the fluid system. A monolayer is formed by the surfactant that causes variation in the surface tension or IFT [94]. Adsorption of NPs depends on the type of the surfactant (anionic, cationic, and non-ionic) and the effective surface charges of the solution (positive or negative). The adsorbed NPs alters the surface charges of the solution, which enhances other interactions like repulsion in the interfacial layers [95]. The distribution of surfactant and NPs at the interface of the solution is arranged in a way that the surfactant monolayer concentrates at the liquid-liquid interface while the counter-ions are beneath the monolayer in the diffuse layer [96]. The surface of the monolayer and the diffuse layer are separated by a surface, which is assumed to be electroneutral [97,98]. The electroneutrality of the bulk phase and interface allows the re-ordering of ions due to the electrostatic effect and the change in entropy. Electroneutrality is the summation of the ions of surfactant adsorbed at the interface and the diffuse counter-ions, which are not uniformly distributed. Based on the electroneutrality of the separating layer of the interface and the diffuse region, both the ionic solution and the surfactant have an effect on the IFT of the liquid-liquid interactions. Prosser and Franses [96] summarized the surface tension of a solution of surfactant and electrolytes,  $\gamma$ , in terms of the interaction forces as follows:

$$\gamma = \gamma_0 + \gamma_{ideal} + \gamma_{cohesive} + \gamma_{electrostatic} \quad (2)$$

where  $\gamma_0$  is the surface tension of the solvent,  $\gamma_{ideal}$  is the ideal contribution (Langmuir) to the surface tension by the monolayer,  $\gamma_{cohesive}$  is the non-ideal contribution (Frumkin) to the surface tension due to the lateral cohesive interaction, and  $\gamma_{electrostatic}$  is the contribution (Davies) to the surface tension by the electrostatics interaction or the 'Maxwell stress' term [99].

Fig. 8 illustrates the IFT (hollow shapes) and zeta potential, ZP, (solid shapes) of different solutions of surfactants with an increase in the concentration of surfactants. In other words, Fig. 8 analyzes the

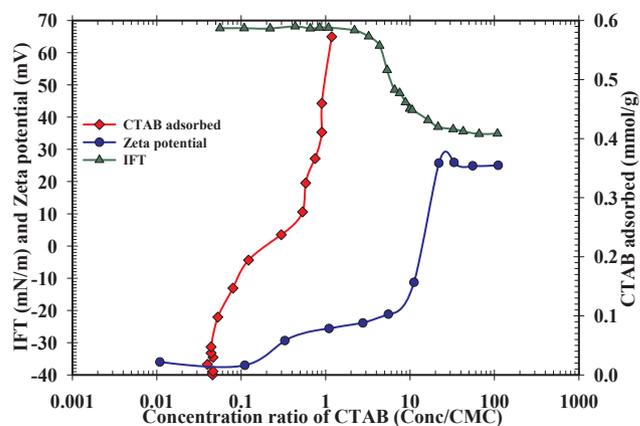


**Fig. 8.** The IFT (hollow shapes) and zeta potential, ZP, (solid shapes) of different solutions of surfactants with an increase in the concentration of surfactants. The circles refer to the experimental data for the solutions of silica NPs and CTAB surfactant reported by Binks et al. [90]. The triangles correspond to the experimental data for the solutions of silica NPs and SDS surfactant reported by Ahualli et al. [87]. The squares and diamonds exhibit the experimental data for the solutions of SDS and SDS with NaCl, respectively, reported by Prosser and Franses [96].

interaction forces in a solution of surfactant-NPs, defined in Eq. (2). The hollow and solid squares in Fig. 8 represent the IFT and the zeta potential for the solutions of the sodium dodecyl sulphate (SDS) surfactant, respectively, reported by Prosser and Franses [96]. The surface tension remains constant but the zeta potential decreases with an increase in the concentration of SDS until  $0.2 \text{ mMol/dm}^3$  is reached. The surface charge of the solution is dominated by the negative ions, which increase with an increase in the concentration of the SDS. However, the balance among the negative ions does not affect the interaction forces that alter the surface tension of the solution. For the concentrations of SDS larger than  $0.2 \text{ mMol/dm}^3$ , the zeta potential increases while the surface tension of the solution decreases with an increase in the concentration of SDS up to  $0.4 \text{ mMol/dm}^3$ . The change in zeta potential is attributed to the formation of hemimicelles by the SDS, which increase the amount of positive ions present in the solution. The surfactant tails (sulphate ions) are arranged inward in a way that the micelle heads are positive. This also depends on the effective charges of the surfactant and the NPs dispersed in solution. The zeta potential and the surface tension remain constant for the concentrations of SDS higher than  $0.4 \text{ mMol/dm}^3$ . This can be described by the domination of the solution by the positive charges of micelles, which prevents further interaction of other forces.

The hollow and solid triangles in Fig. 8 represent the IFT and the zeta potential for the solutions of silica NPs and SDS, respectively, reported by Ahualli et al. [87]. It is revealed that the water-octadecene IFT decreases with an increase in the concentration of SDS at the certain concentration of silica NPs [87]. However, the zeta potential remains constant with an increase in the concentration of SDS until  $0.2 \text{ mMol/dm}^3$ . It is inferred that the negative ions of the SDS are balanced by the ions of the NPs. The interaction forces lead to a reduction in the IFT but no change in the zeta potential. The zeta potential of the solution decreases with an increase in the concentration of SDS larger than  $0.2 \text{ mMol/dm}^3$ . The negative ions of the SDS increase with an increase in the concentration of SDS, which causes a further reduction in the IFT. Ahualli et al. [87] separated the excess surfactant from the silica NPs dispersed in solution before measuring the zeta potential, as shown in Fig. 8. The same phenomenon was observed in the IFT of the solutions of NaCl and SDS (hollow diamonds) but the value of the IFT is smaller than SDS alone (hollow squares).

The hollow and solid circles in Fig. 8 represent the IFT and the zeta



**Fig. 9.** The IFT and the zeta potential (left vertical axis) and the CTAB adsorbed (right vertical axis) versus the concentration ratio of CTAB. The experimental data used here are extracted from the work conducted by Binks et al. [90].

potential for the solutions of silica NPs and cetyl trimethyl ammonium bromide (CTAB) surfactant, respectively, reported by Binks et al. [90]. The zeta potential of the solutions of CTAB and silica NPs shows the similar behavior to the solutions of SDS and silica NPs. However, the zeta potential of the solutions of CTAB and silica NPs increases due to an increase in the concentration of CTAB, as shown in Fig. 8. The IFT of the solutions remains constant with an increase in the concentration of CTAB up to  $4 \text{ mMol/dm}^3$ . This is because of the attraction between the silica NPs and the CTAB ions, which prevents the ions from reducing the surface tension. The increase in the zeta potential is due to the formations of hemimicelles and micelles.

The surface of silica NPs is coated with CTAB when a low concentration of CTAB is added to the solution to enhance its stability [100]. A low concentration of CTAB prevents the aggregation of silica NPs [101] depending on the length of the alkyl chains [100]. Therefore, the initial surfactant concentration does not alter the IFT but ensures the stability of the NPs through the steric repulsion. Fig. 9 shows the IFT and the zeta potential (left vertical axis) and the CTAB adsorbed (right vertical axis) versus the concentration ratio of CTAB using the experimental data extracted from the work conducted by Binks et al. [90]. The CTAB adsorbed increases with an increase in the concentration of CTAB above the CMC. In addition, the zeta potential increases while the IFT decreases. However, the IFT of the solutions of CTAB and silica NPs is larger than the IFT of CTAB alone.

Increasing the surfactant concentration enhances its adsorption (attraction) to the oppositely charged NPs, which makes the zeta potential to approach  $0 \text{ mV}$  resulting in aggregation [100]. Repulsion of the surfactant coated NPs increases with an increase in the surfactant concentration. As a result, the aggregation of the NPs becomes reverse, which refers to the peptization, when the concentration of CTAB increases [100]. The surfactant adsorption increases with an increase in the concentration of surfactant due to the reordering pattern of the surfactant tail-head and head-tail at the interface, which results in the repulsion and attraction of NPs in order to accommodate more surfactant [100].

Repulsion of the surfactant and NPs of similar charges enhances the stability of a solution of NPs at a low concentration of surfactant. The process of surfactant adsorption in the solutions of surfactant and NPs with the same charges is similar to that in the solutions of surfactant and NPs with the opposite charges. Therefore, the IFT decreases and the zeta potential changes based on the surfactant charges [101–103].

### 3.3. Cationic surfactant and NPs

The addition of a cationic surfactant to the solution of anionic (negatively charged surface) NPs reduces the effective surface charges

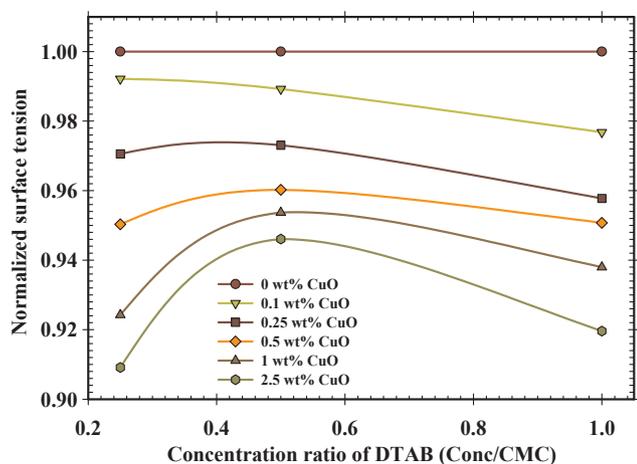


Fig. 10. Normalized surface tension of water at different concentrations of CuO NPs versus the concentration ratio of DTAB. The experimental data used here were reported by Harikrishnan et al. [94].

of the solution from negative towards positive but the interaction radius between the NPs and the surfactant remains constant with an increase in the concentration of surfactant [87]. This results in the aggregation of the particles, as discussed in Section 3.2. The oil-water IFT of a solution of anionic NPs and cationic surfactant is larger than the IFT of cationic surfactant without NPs. This can be attributed to the electrical attraction between the NPs and the surfactant, which reduces the amount of ions in the solution [87]. However, Fereidooni and Azizian [101] observed that when a low concentration of CTAB is added to ZnO NPs dispersed in deionized water, which is slightly negative charged [15], the oil-water IFT is reduced. A similar behavior was observed for the air-water surface tension at a low concentration of solution of CuO NPs and dodecyl trimethyl ammonium bromide (DTAB), as shown in Fig. 10. Increasing the concentration of the NPs at constant surfactant concentration reduces the surface tension.

However, different behavior patterns are obtained for the surface tension at a constant concentration of NPs with an increase in the surfactant concentration, as shown in Figs. 10–12. It is revealed from Fig. 10 that at low concentration of NPs (0.1 wt%), the surface tension reduces with an increase in the concentration of the surfactant [104]. For concentrations of NPs larger than 0.5 wt%, the surface tension increases with an increase in the surfactant concentration and it reaches a

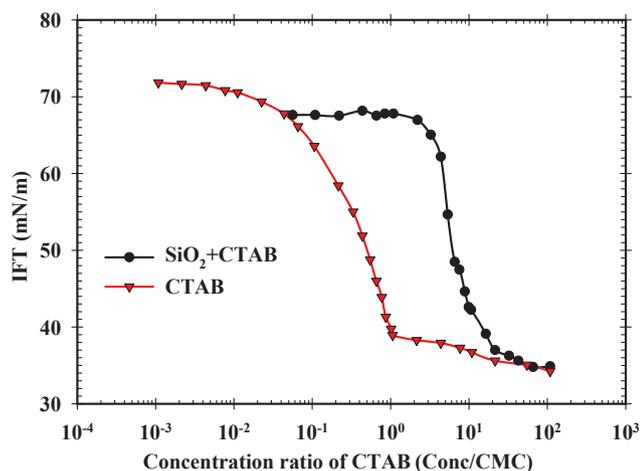


Fig. 11. The IFT of solutions of CTAB surfactant with (black circles) and without silica NPs (red triangles) versus the concentration ratio of CTAB. The experimental data used here were reported by Binks et al. [90]. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

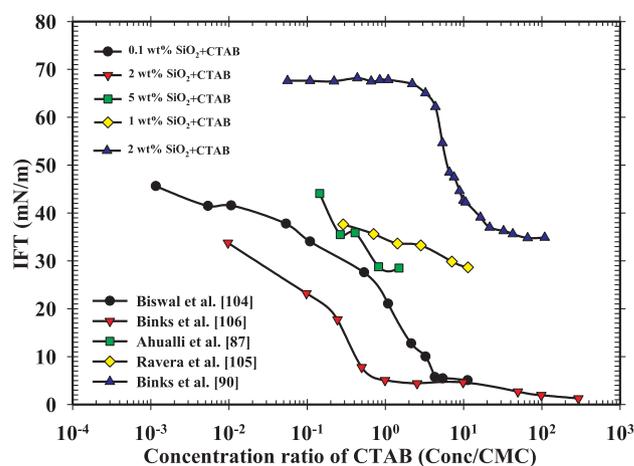


Fig. 12. The IFT of different solutions of silica NPs and CTAB against the concentration ratio of CTAB. The experimental data used here were reported in literature [87,90,104–106].

maximum value at the surfactant concentration equal to 0.5 CMC. Then, the surface tension reduces and it approaches its initial value at the surfactant concentration equal to 0.2 CMC [90].

### 3.4. Anionic surfactant and NPs

The effective charges of a dispersed solution of anionic NPs increase with the addition of anionic surfactant. This is due to the repulsion between the ions of NPs and the surfactant adsorbed [87]. The IFT and the surface tension of the solutions of surfactant and NPs are smaller than the solutions of surfactant without NPs for oil-water and air-water systems, respectively. The migration of surfactant molecules from the bulk phase to the interface causes the repulsion of the opposite charges [104]. Ma et al. [107] explained that the repulsive Coulomb interactions between the surfactant and the NPs enhance the diffusion of surfactant towards the interface, which accounts for the reduction in IFT. The ions of the NPs and the surfactant deplete each other to balance the repulsive forces, which results in the negative free energy of the surfactant adsorption and micelle formation [107]. The free energy of surfactant at the interface is smaller than that in the bulk phase. Thus, less work is required by the ions at the interface, which leads to a reduction in the value of IFT [108].

Figs. 13 and 14 show the normalized surface tension, which is defined as the ratio of the surface tension of the solution of surfactant and

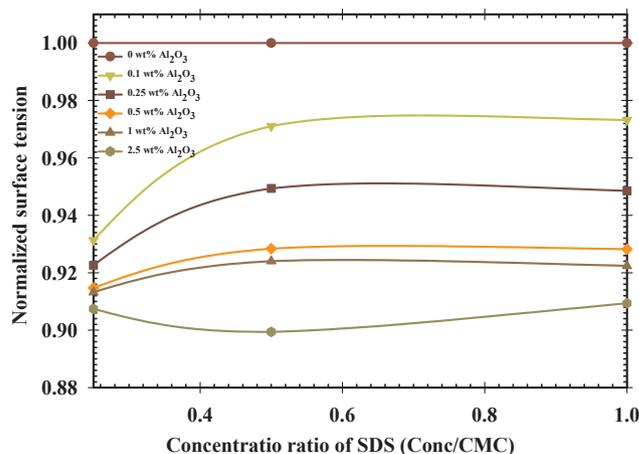


Fig. 13. Normalized surface tension of water at different concentrations of Al<sub>2</sub>O<sub>3</sub> NPs versus the concentration ratio of SDS. The experimental data used here were reported by Harikrishnan et al. [94].

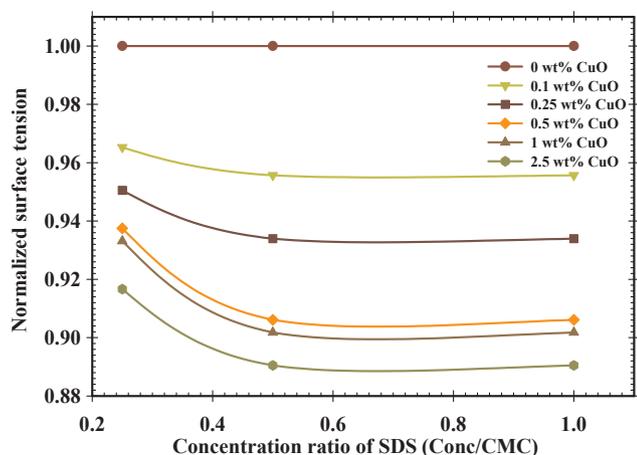


Fig. 14. Normalized surface tension of water at different concentrations of CuO NPs versus the concentration ratio of SDS. The experimental data used here were reported by Harikrishnan et al. [94].

NPs at a certain concentration of surfactant to the surface tension of the solution of only surfactant, versus the concentration of an anionic surfactant at different concentrations of NPs. The surface tension of the solutions of Al<sub>2</sub>O<sub>3</sub> NPs and SDS surfactant (Fig. 13) increases with an increase in the concentration of surfactant when the concentration of Al<sub>2</sub>O<sub>3</sub> NPs is constant except for the solution with 2.5 wt% concentration of Al<sub>2</sub>O<sub>3</sub> NPs. The behavior of the surface tension of the solutions of SDS and Al<sub>2</sub>O<sub>3</sub> NPs is similar to that of DTAB and silica NPs in Fig. 11.

The behavior of the surface tension is different for the solutions of NPs and surfactant, which have the same charges. The surface tension reduces with an increase in the concentration of either surfactant or NPs until the surface tension reaches an equilibrium value. This explains that other interactive forces exist in the solution alongside the steric repulsion and the electrostatic forces between the head groups, the alkyl chains, and the particles [96], which result in the variation of IFT, as shown in Fig. 15. The observation of the behavior of IFT for the same charged NPs and surfactant shows that the IFT increases when the concentration of surfactant is larger than CMC. Dong and Johnson [109] explained that the capillary force is responsible for the increase in the IFT, which is observed at a surfactant concentration close to CMC. The deformation of the interface by the random displacement of the particles causes the change in effective surface tension.

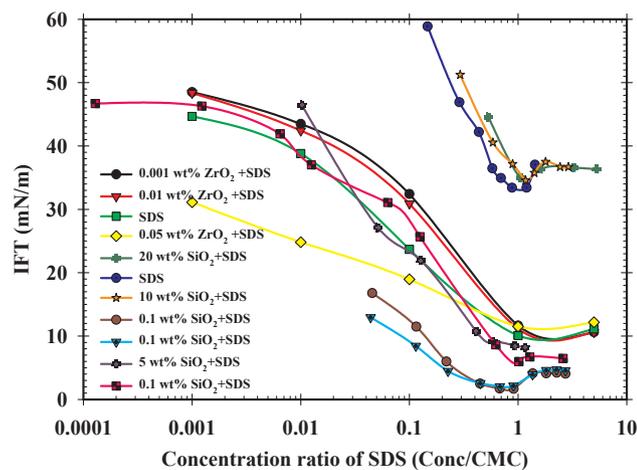


Fig. 15. The IFT of different solutions of negatively charged NPs and anionic surfactant versus the concentrations ratio of SDS. The experimental data used here were reported in literature [27,55,87,107].

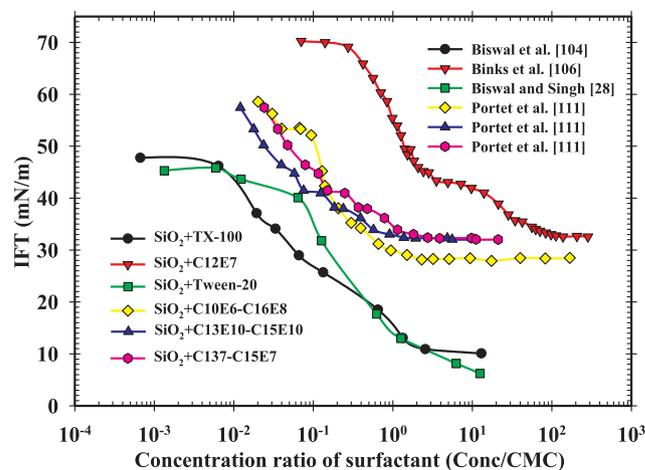


Fig. 16. The IFT of different solutions of non-ionic surfactants and silica NPs versus the concentration ratio of surfactants. The experimental data used here were reported in literature [28,104,106,111].

### 3.5. Non-ionic surfactant and NPs

Non-ionic surfactants are different from other types of surfactants because the surface tension is rarely impacted by the effective charges of the anionic or cationic NPs. The interaction radius shows that there is no relationship between the surfactant and NPs [107]. This is attributed to the weak interaction among the effective charges in the solution and the size of NPs. The surfactant is not adsorbed at the liquid-liquid interface; instead, it remains in the bulk phase of one of the liquids [107]. The IFT of non-ionic surfactant in the presence of NPs varies with the concentration of the surfactant and the effective charges of the NPs. Biswal et al. [104] explained that the effect of NPs at low surfactant concentrations is negligible but become pronounced at high concentrations of surfactant, as shown in Fig. 16. The migration of surfactant from the bulk phase to the interface is retarded, which results in an increase in the IFT [28]. Adsorption isotherm of solutions of non-ionic surfactant and NPs is similar to the other surfactants, but the variation in IFT is different. This behavior is attributed to hydrolysis, DLVO, and non DLVO forces occurring in the process [96,110].

### 4. Low salinity water and surfactant

It has been shown that salinity is an important factor in the phase behavior of surfactant-oil-water micro-emulsion [35,112,113]. An optimum salinity of the solution is important for effective oil recovery. Teklu et al. [114] explained that several mechanisms account for the effective oil recovery during the injection of LSW in combination with the surfactant. Also, the minerals in the reservoir rock and the type of reservoir formation play important roles in the recovery mechanisms [115,116]. Some of the mechanisms responsible for the oil recovery by combination of LSW and surfactant are the osmosis, the capillary pressure, the diffusion, the wettability alteration, the reduction in IFT, and other relevant forces like EDL effect. The osmotic forces result from the capillary force or chemical potential gradient of the salt. Imbibition of LSW into the porous medium through osmotic capillary force drives the oil droplet out of the mesopores and macro-pores [114].

However, there are limited studies available on the effects of combination of LSW and surfactant on the oil recovery. Injection of LSW (as chase fluid) prior to surfactant injection can improve the recovery factor through alteration of the wettability and reduction in the IFT [46]. Increasing the salinity of the injected water increases the contact angle, which results in lower oil recovery, but reduces the IFT, as shown in Fig. 17 [117]. Also, the valence of the ions present in the brine affects the IFT of the system [118]. More studies are required to evaluate the

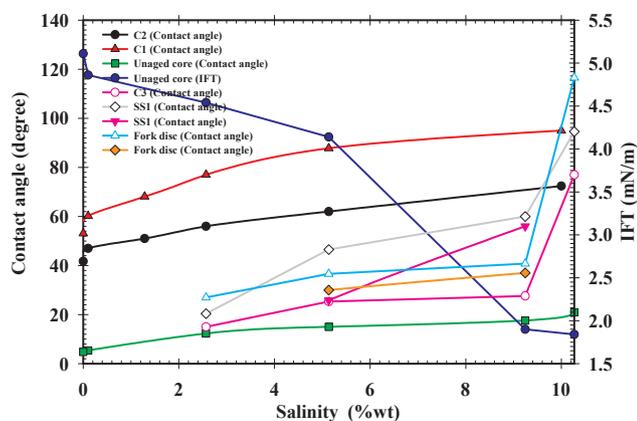


Fig. 17. The contact angle and IFT versus the salinity of the solution at a constant concentration of surfactant, where 'C' represents carbonate and 'SS' represents sandstone. The numbers 1, 2, and 3 stand for different core rocks. The solid shapes refer to solutions of LSW and surfactant while the hollow shapes correspond to LSW. The experimental data are extracted from the works conducted by Alameri et al. [46] and Teklu et al. [47].

effects of LSW alternating surfactant injection on the IFT and the contact angle during EOR. Consequently, an optimal salinity for LSW injection and an optimal concentration of surfactant are critical to optimize the oil recovery.

## 5. Summary and conclusions

The mechanisms involved during oil recovery from sandstone and carbonate reservoirs using combination of LSW, surfactant, and NPs are more complicated for EOR. Some of these mechanisms are the multi-ion exchange, the disjoining pressure, the electrophoresis, the adsorption, the diffusion, the fines migration, the osmotic pressure, and the EDL effects. Several factors which contribute to the mechanisms are listed as follows:

- The effective charges of the dispersed solution of NPs,
- The effective charges of the LSW,
- The type of surfactant,
- The concentration of surfactant,
- The concentration of NPs in solution (volume fraction),
- The valence electrons of the ions of LSW,
- The size of the NPs, and
- The minerals in the reservoir rock.

Therefore, the following conclusions can be extracted from this study:

- The ability of NPs to reduce fines migration during the injection of LSW into the reservoir depends on the type of reservoir, the composition of the brine, and the effective charges of the NPs in solution.
- Increasing the concentration of NPs in deionized water and low and high salinity water results in the cyclic variation of the contact angle as well as the recovery factor. The variation in the recovery is attributed to the interactions between the contact force and the disjoining pressure of the NPs in the system.
- Surfactant enhances the stability of NPs in solution and reduces the IFT. The behavior is due to the effective charges of the ions of NPs and surfactant. Also, the structure of the micelles formed at concentrations larger than CMC directly affects the IFT.
- The IFT of solutions of LSW and surfactant decreases while the contact angle increases by increasing the salinity of the solution.

Despite the successful implementation of LSW and surfactant for EOR, special attention needs to be painstakingly considered during the selection of a desired combination of LSW, surfactant, and NPs. This comprehensive review on the variation in contact angle, the stability of NPs, and the IFT during injection of different combinations of LSW, surfactant, and NPs can be used for successful implementation of EOR in sandstone and carbonate reservoirs.

## Acknowledgment

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