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Applications of Surfactants and Nanoparticles in Enhanced Oil Recovery Processes

Christian A. Paternina

Abstract

The surfactant injection is considered as the EOR (Enhanced Oil Recovery) with the highest potential to recover oil from reservoirs due to its ability to reduce interfacial forces into the porous medium. However, the adsorption of this type of chemical on the surface of rocks is the main problem when a surfactant injection project is applied since the surfactant molecules would rather be placed on rock minerals instead of being the oil–water interface. Based on this fact, this chapter would be discussed the significance of surfactant injection as an EOR method, the types of surfactants used, the main mechanism and parameters involved in the surfactant adsorption on the rock, and its consequences in oil recovery. Likewise, the addition of nanoparticles to inhibit the adsorption of surfactants is another topic that will be covered as a novel technology to improve the efficiency of the EOR process.

Keywords: Surfactants, Adsorption, EOR, Rock, Porous medium, nanoparticles

1. Introduction

In the hydrocarbon industry, there is a constant need to obtain large volumes of oil, which leads to the development of new techniques to extract the greater amount of oil into the reservoirs, which, using conventional technologies, would be impossible to achieve. The application of Enhanced Oil Recovery or EOR methods has been an opportunity to accomplish a substantial increase in hydrocarbon production in depleted reservoirs.

The surfactant injection is a recovery technique with great potential for recovery [1], which has shown an important effect in increasing the capillary number “ N_c ” (dimensionless variable, which relates viscous with interfacial forces). However, due to the great number of difficulties of its application in field, the efficiency of the process in many cases is very low. Among the most notable problems in the application of this method, it is the adsorption of surfactant on the rock, which leads to the chemical being trapped on the surface of the mineral substrate instead being positioned at the water–oil interface to reduce the interfacial tension.

2. Surfactant injection

Among the recovery methods, the surfactant injection has been listed as one with the greatest potential. It has been implemented in China, the United States,

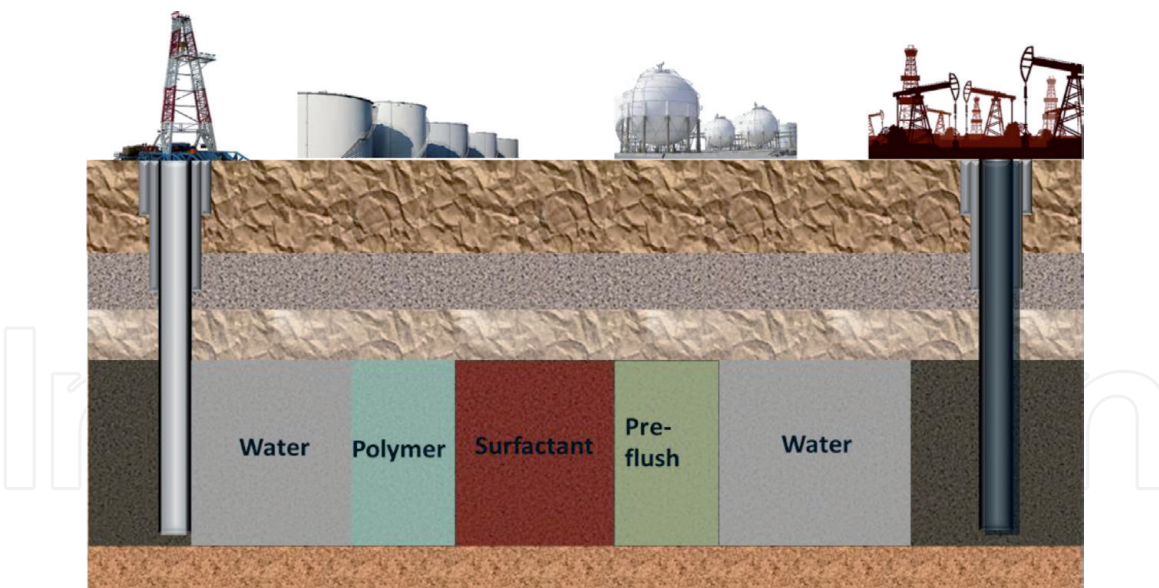


Figure 1.
Surfactant injection process.

France, Austria, Oman and Canada [2]; throughout history has achieved recovery factors up to 60% of the OOIP (Original Oil in Place) [3]. Due to the behavior of surfactants to reduce the interfacial tension at the oil–water interface to ultra-low values of up to 10^{-3} dynes/cm, easing the mobilization of the oil bank between 10% - 20% of the oil remaining in the formation, which was trapped in the porous medium. Successively, it can be pushed by a polymeric solution, which improves the mobility relationship between water and oil, favoring the final recovery factor achieved through the tertiary recovery technique [4].

A typical surfactant injection process is composed of different fluid stages or bumps, which can be distinguished in **Figure 1**. Thus, the first fluid injected into the formation consists of a fresh water pre-flush, which Its objective is to adjust or reduce the salinity of the formation, to avoid that the high salinity present in most oil fields, favors the precipitation or the adsorption of the surfactant to be injected; then, a surfactant formulation is displaced to transport the surfactants capable of positioning themselves between the oil–water phases and reducing the interfacial tension, favoring their movement to the surface. Afterwards, it is recommended to inject a bump of a high viscous polymer solution, intended to increase the viscosity of the aqueous solution and improve the sweep efficiency in the formation. Finally, water is injected to displace previously injected fluids.

2.1 Surfactant composition

Surfactants are amphiphilic molecules, which have a hydrophobic (non polar) part known as the “tail” and another hydrophilic (polar) called “head”, as can be seen in **Figure 2**. The tail is made up of non polar groups (hydrocarbons and/or fluorocarbons) and the head, is composed of ions and/or polar compounds such as sulfates, sulfonates, carboxylate, phosphates or quaternary ammonium [5]. Surfactants due to their dual affinity nature, both for the hydrocarbon and the aqueous phase, can be used to improve recovery processes by reducing the free energy of the water–oil interface into reservoirs; resulting in an improved of the microscopic displacement efficiency [6].

The interfacial tension reduction achieved by surfactants is because of the adsorption of surfactant monomers in the interfacial region (water–oil) [7] and this fact makes the surfactant a versatile chemical which is widely used in different

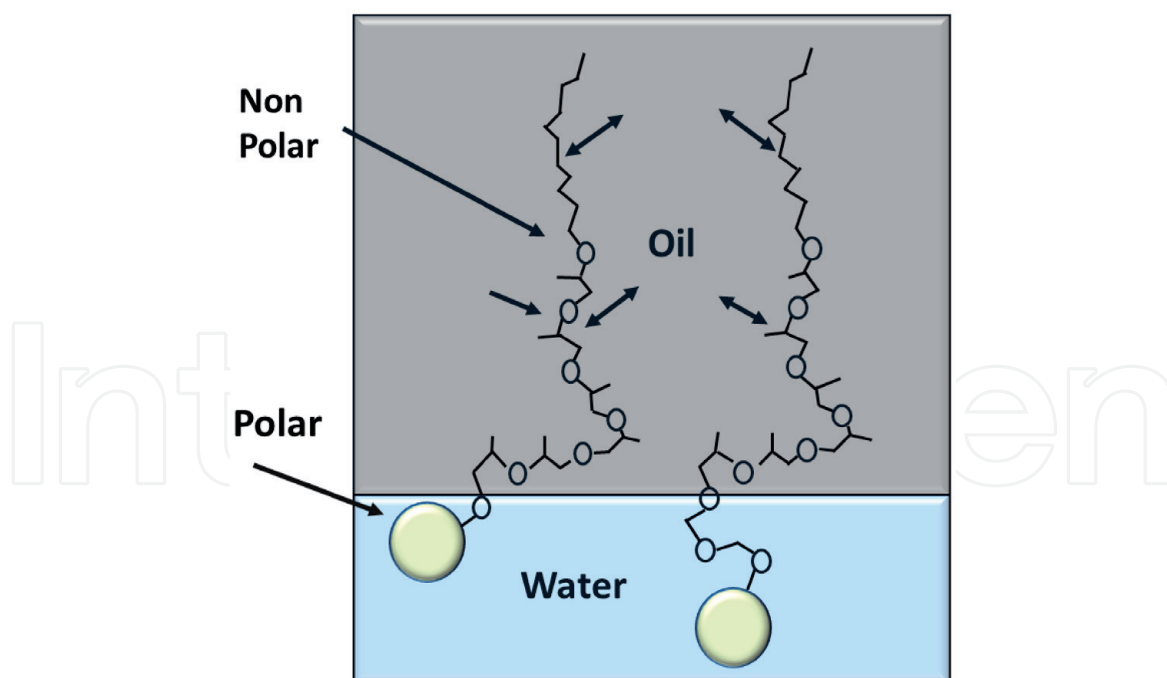


Figure 2.
 Molecular structure of a surfactant.

kinds of processes in the oil industry such as: the formation of foams and emulsions, stabilization of fines, modification or change of wettability and of course enhanced oil recovery.

2.2 Types of surfactants

Surfactants are highly complex compounds used in a wide variety of industries such as pharmaceuticals, cosmetics, food, agrochemicals, and oil among others. In the hydrocarbon industry, surfactants have different uses among which can be mentioned: (de) emulsifiers, (anti) foaming agents, corrosion inhibitors, dispersants, humectants. Thus, there have been different forms to classify the surfactants, however, the most used what makes a distinction of the head nature head as shown below [8].

a. Anionic Surfactants:

Anionic surfactants have negatively charged hydrophilic group, consisting mainly of a sulfate, a sulfonate, a carboxylate or a phosphate. These surfactants dissociate into an amphiphilic ion and a cation, consisting of an alkali metal (Na^+ , K^+) or a quaternary ammonium, as is the case of alkylbenzene sulfonate ($\text{R-C}_6\text{H}_4\text{SO}_3^- \text{Na}^+$) [9]. Anionic surfactants are commonly used in enhanced oil recovery processes, since, thanks to its negative charge in the polar head, it is repelled by the clays and sandstones from the reservoirs, reducing its retention in the porous medium.

b. Cationic Surfactants:

Cationic surfactants have a positive charge on their polar head, usually made up of ammonium, pyridinium, or quaternary ammonium. When these surfactants are dissociated forming a cation and a halogen-type ion (Cl^-). One of the most used cationic surfactants is quaternary amine chloride

($\text{R-N(CH}_3)_3^+\text{Cl}^-$). Regarding their applications in EOR, they are used mainly in carbonate formations, in order to reduce their adsorption, since the surface charge of limestone and dolomites is positive, and they repel surfactants of the same charge.

c. Non-ionic surfactants:

They are those surfactants with their polar part soluble in water, thanks to the inclusion of oxygen or similar atoms such as polyethylene glycol. However, they do not have an electric charge and when they dissociate in water, there is no change in the electric force, for example polyoxyethylated alkylphenol ($\text{R-C}_6\text{H}_4(\text{OC}_2\text{H}_4)_x\text{-OH}$).

d. Amphoteric or Zwitterionic Surfactants:

Amphoteric surfactants possess both charges when dissociating in aqueous medium, depending on pH. Thus, they behave as cationic surfactants in acid solution and anionic in a basic medium. Among the chemical structures that are part of the polar head of the molecule are the compounds of amine oxide, betaine and carboxylated amines [10].

e. Gemini surfactants:

Gemini surfactants are a new family of surfactants that have at least two hydrocarbon tails and two polar or ionic groups, with a great variety of spacers of different nature, among which stand out short or long methylene groups, rigid groups (stilbene), polar (polyether) and nonpolar (aliphatic, aromatic) compounds. The ionic group can be positive (ammonium) or negative (phosphate, sulfate, carboxylate), while the polar non-ionic ones can be polyether or sugar [11].

The great advantage of Gemini surfactants over conventional surfactants is that they have a low CMC (Critical Micellar Concentration), high surface activity, better stability, high tolerance to hardness and can be used in low permeability reservoirs [12].

f. Biosurfactants:

Biosurfactants is a wide variety of amphiphilic molecules synthesized by plants, animal, and microbes. These molecules have a large range of molecular weight and are environmentally safe, making this kind of surfactants noteworthy for applications in oil industry and in the recent years in Enhanced Oil Recovery process specifically [13]. The use of biosurfactants have been applied in MEOR (Microbial in Enhanced Oil Recovery) since they have shown equal or better behavior than its chemical counterpart in several parameters especially for environmental compatibility [14]. However, this type of technology needs a large amount of resources and investment to overcome the traditional surfactants, even though the increase of green practices, there is a bright future for biosurfactant in EOR.

In **Figure 3** it is possible to observe the structure of the different types of surfactants according to their classification discussed above.

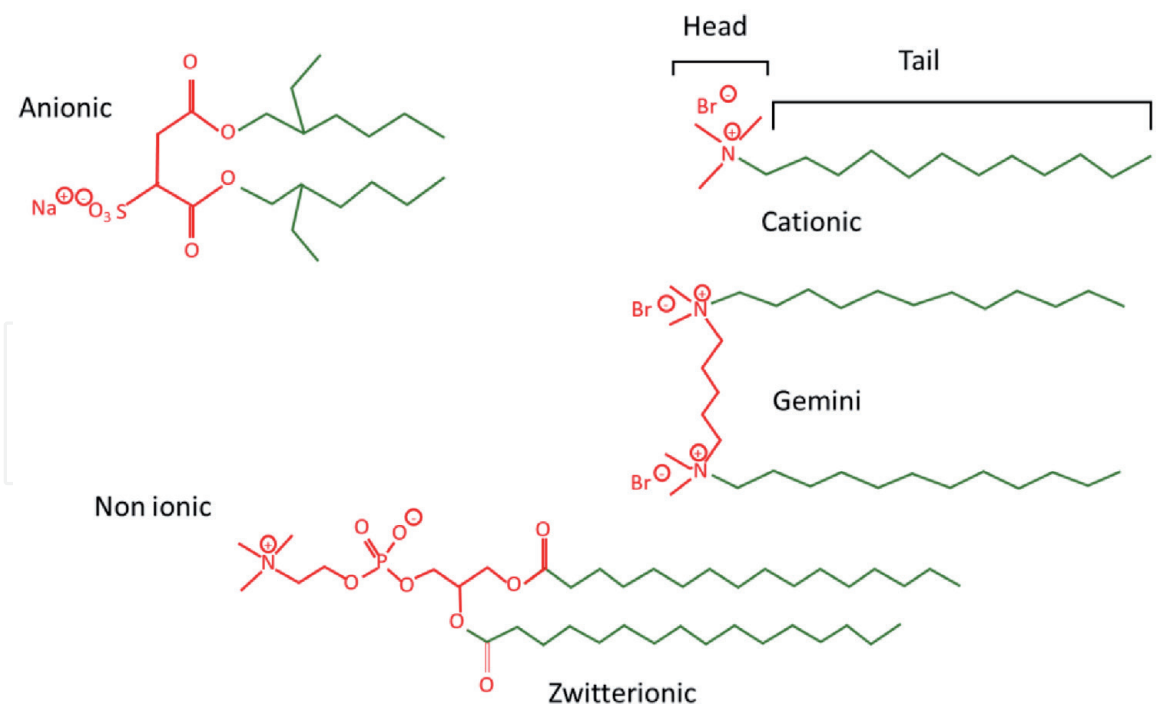


Figure 3.
 Molecular structure of the different types of surfactants [15].

3. Adsorption of surfactants into the porous medium

Despite of the great potential shown by the surfactant injection for oil recovery, this methodology has high costs compared to other methods mainly due to the price of the injected fluids, additionally different problems arise when it comes to its field implementation in both technical and financial.

Among the many drawbacks of this recovery method, it can mention the loss of surfactants by their adsorption on the rocks of the reservoir, which has a large impact on determining whether the process is viable or not; since if the most of the injected chemical is trapped in the formation, it will not be available to locate at the oil–water interface and thus reduce the interfacial tension of the system [16, 17].

The adsorption of surfactants on a solid substrate can be defined as the selective distribution of the adsorbate around the surface of the rock medium, since it is energetically more favorable for the molecules than to be located at the oil–water interface [18]. The interactions of the surfactant molecules with the mineral substrate can be electrostatic or due to hydrophobic interactions as shown in **Figure 4**.

3.1 Surfactant adsorption mechanisms

To the surfactant being adsorbed at the solid–liquid interface, there are a series of mechanisms that cause the surfactant to adhere to the reservoir rock, such as:

a. Ion exchange:

Which involves the displacement or exchange of counterions adsorbed on the substrate by similarly charged ions. For example, a cationic surfactant can displace an adsorbed sodium cation on a clay and thus take its place on the surface of the mineral substrate (**Figure 5a**) [20–22].

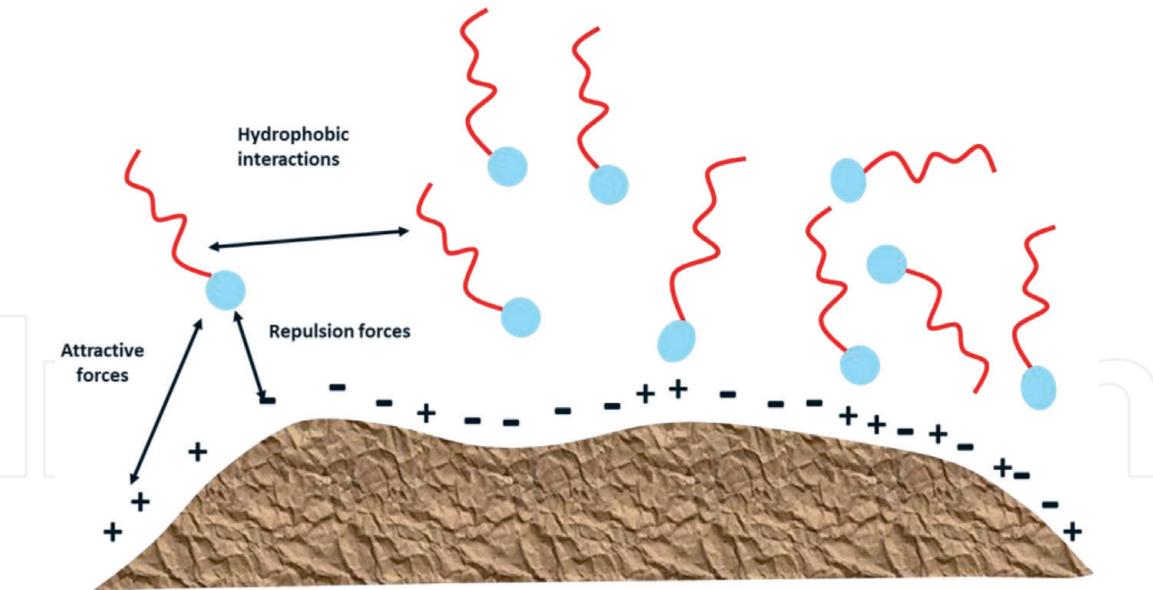


Figure 4.
Adsorption of surfactants on a substrate [19].

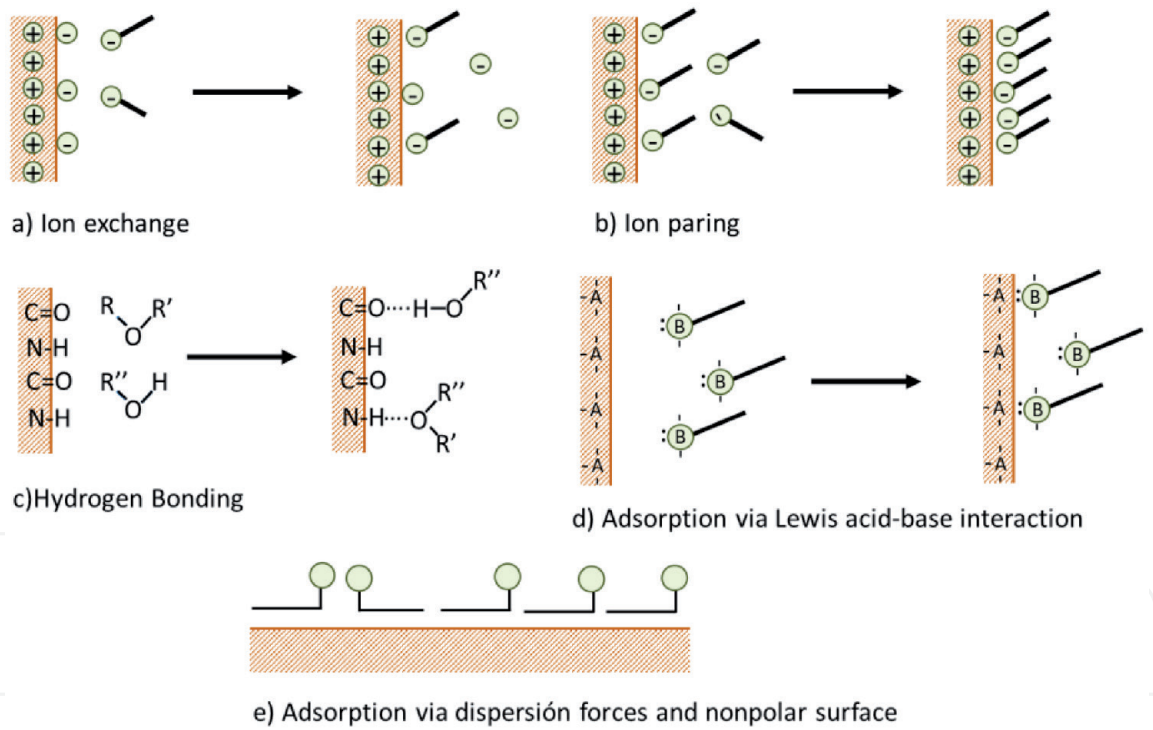


Figure 5.
Mechanisms of surfactant adsorption on solid substrates [5].

b. Ionic Pairing:

The adsorption of ionic surfactants on an oppositely charged solid surface to the surfactant polar head (**Figure 5b**) [21, 22].

c. Acid–Base Interactions:

Adsorption by means of hydrogen bonds (**Figure 5c**) between the substrate and the adsorbate (surfactant) [21–23] or by acid–base Lewis reaction, as seen in **Figure 5d**.

d. Adsorption by dispersion forces:

The London-Van der Waals dispersion forces act between the adsorbent molecules (mineral or solid substrate) and the adsorbate (surfactant) (**Figure 5e**). Adsorption by this type of mechanism generally increases with the molecular weight of the adsorbate and may be accompanied by other types of interactions that assist the adsorption, thus, this mechanism favors the ability of surfactant molecules to displace others from the interface [22, 24].

e. Hydrophobic bonds:

The combination of the attraction given by the hydrophobic groups of the surfactant molecules and their tendency to escape from the aqueous medium in which they are on allow the surfactants to be located on the solid surface due to the aggregation of their hydrocarbon chains [5].

3.2 Surfactant adsorption parameters

The adsorption of surfactants on the rocks from oil reservoirs is a phenomenon that is influenced by a wide number of parameters, however, there are some that produce a better synergy of the chemical with the rock surface, among which are:

a. Surfactant concentration:

It has been shown that, by increasing the concentration of surfactant in the aqueous solution in contact with a substrate, the amount of surfactant absorbed on said surface increases, since there is a greater availability of the chemical to be located in said spaces [2]. Thus, the importance to determinate the surfactant concentration to inject into a reservoir in EOR process, normally to values close to CMC (around 2000 ppm).

b. Temperature:

The increase in temperature leads to a slight decrease in the amount of surfactant adsorbed, due to a substantial increase in the translational kinetic energy and the entropy of the system, which reduces the attractive forces between the surfactant and the reservoir rock, avoiding the formation of an organized layer of surfactant monomers at the solid-liquid interface [2].

c. Hydrogen potential, pH:

The pH of the aqueous medium essentially influences the adsorption, because rock surfaces are positively or negatively charged, depending on the degree of dissociation of functional groups on their surface, so a change in pH alters the surface charge of minerals [25]. Thus, the amount of surfactant adsorbed on a solid can be altered by changing the pH of the medium. Therefore, in general, in the case of anionic surfactants (with a negative charge), adsorption decreases at low pH and for cationic surfactants (positive charge) it increases with pH [26].

d. Salinity:

The presence of salts in the reservoir affects the solubility and the formation of more complex aggregates formed by the surfactant molecules [27] due to the

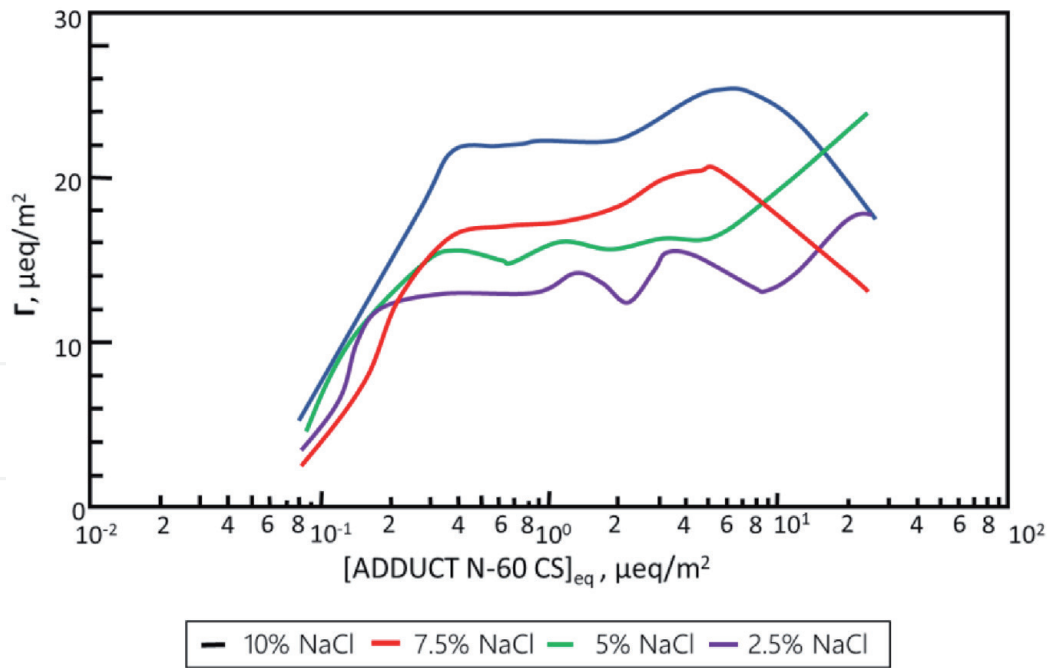


Figure 6.
Effect of NaCl concentration on the adsorption of an anionic surfactant [34].

electrostatic interactions of “salting-in” and “salting-out.” Where salts act as facilitators of aggregate formation [28, 29]. Additionally, when a compression of the so-called electrical double layer occurs [30, 31], the adsorption density of ionic surfactants at the interface is modified and the structure and morphology of the layers of micellar aggregates is changed [32]. On the other hand, the presence of electrolytes decreases the CMC (Critical Micellar Concentration) since it produce the decreasing of the repulsive forces between the surfactant molecules [19, 33].

Figure 6, it is possible to see the study case developed by Figdore [34], where it is described the adsorption of an anionic surfactant on a sandstone substrate at different NaCl concentrations in the aqueous medium [34]. Adsorption isotherms are observed, where it is possible to observe that meanwhile salt concentration in the medium increases, the adsorption of the surfactant also increases proportionally. This is attributed to the reduction of the repulsive forces between the negative heads of the surfactants and the surface of the solid substrate (also negatively charged), due to the electrolytes (Na^+), allowing the surfactant molecules to adhere with easier on the surface of the sand grains.

a. Mineralogy:

It is of great importance to identify the minerals that make up the solid substrate or porous medium, which will be contacted by the injected surfactants since depending on which components form the surface of the rock, certain adsorption mechanisms exist and will give a guideline for thus select the type of surfactants that should be implemented in an enhanced recovery processes to reduce the chemical adsorption on the rock. Typically, surfactants with the same charge to the substrate of interest are selected to reduce the magnitude of adsorption [35].

Examples of common mineralogies in oil and gas reservoirs are quartz sandstones and clay minerals such as kaolinite.

- Kaolinite:

It is formed by a clay mineral with the chemical formula $\text{Al}_2(\text{OH})_4\text{Si}_2\text{O}_5$, which is a claystone, with a laminar structure, made up of a tetrahedral sheet of silica and another, octahedral of alumina [7]. At neutral pH, kaolinite is negatively charged on the faces of its surface and positively on the edges of the same. The isoelectric point of kaolinite occurs at low pH, but it is at pH values close to or greater than 4.6, the negative charge density increases significantly on its surface [36]. Additionally, the level of adsorption of surfactants on kaolinite is higher compared to other porous solids since it has a large surface area [37].

- Sandstone:

It is a type of rock, in which silica prevails. This compound acquires a surface charge depending on the relative concentration of H^+ and OH^- in solution; Therefore, the charge of this surface depends mainly on the pH. On the other hand, the isoelectric point of silica occurs approximately at a pH of 2, showing that the negative charge density remains low, until the pH reaches values higher than 6 [38].

Other parameter that has to be kept on mind is the use of the thermodynamic potential of the Gibbs free energy, which relates the variations of the enthalpy and entropy of a system, indicating the spontaneity of the reaction and in our case of the adsorption. Thus, the greater the negative of the Gibbs energy variation, the process will be energetically favored.

4. Influence of nanoparticles on surfactant adsorption

Due to the inconveniences experienced by the injection of surfactants, in which there are a low reduction in interfacial tension, high technical costs and large amounts of surfactant adsorbed on the rock; new technologies have been proposed to optimize surfactant injection processes, where the application of nanoparticles is seen as a proposal to improve the technique performance.

Broadly speaking, the application of nanoparticles in EOR processes can be divided into three main types: nanofluids, nanoemulsions and nanocatalysts. The first two have great applicability in surfactant recovery techniques since they seek to influence capillary forces to obtain an increase in the recovery factor. The EOR mechanisms of nanofluids have already been investigated in literatures, which mainly includes disjoining pressure, pore channels plugging, viscosity increase of injection fluids, IFT reduction, wettability alteration and preventing asphaltene precipitation. On the other hand, the use of nanocatalysts has shown great applicability in thermal recovery processes, since nanoparticles allow an accelerated decrease in the viscosity of crude oil [39]. However, the level of viscosity reduction is related to the types of nanoparticles, their concentration, and fluid temperature.

There are different studies that affirm that the addition of nanoparticles to a solution with surfactants can be beneficial for increasing the recovery factor [40–42]. However, the predominant mechanisms that favor the increase in oil recovery have not been determined with certainty. Reduction on the adsorption of surfactants on the rock surface could be a powerful mechanism to improve recovery processes. However, the number of studies of this type has been limited. However, in the existing studies of this topic, it has been shown that nanoparticles produce a reduction in the amount of surfactant adsorbed on the rock [43–48] as can be seen in **Figure 7**, meanwhile the nanoparticle of Silica oxide concentration increases the amount of adsorbed SDS (Sodium dodecyl sulfate) surfactant decreases.

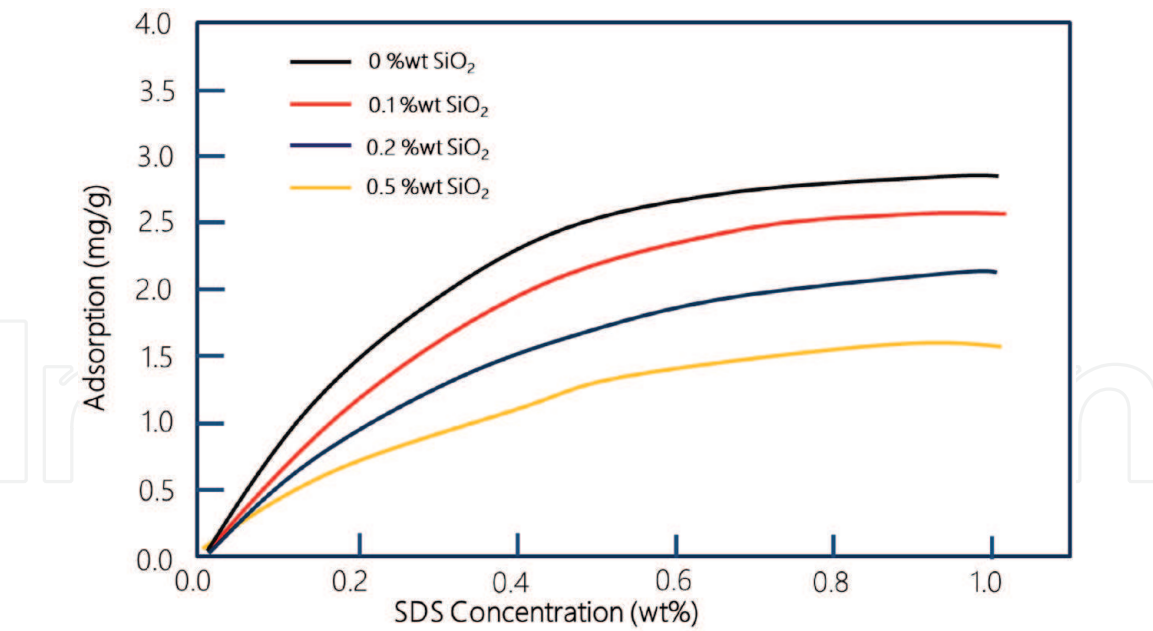


Figure 7.
Effect of the addition of silica oxide nanoparticles on the adsorption of SDS surfactant [45].

In the Zargartalebi study of 2014, two types of silica nanoparticles of different hydrophobicity were implemented with an anionic surfactant SDS (sodium Dodecyl Sulfate) on a substrate to determine the effect on the adsorption of the surfactant. The results of the experiment showed that indeed in both cases there was a reduction in the magnitude of the adsorption in general at lower values of the CMC of the surfactant used. It was observed that the hydrophobic nanoparticle showed a greater reduction in the amount of surfactant adsorbed due to the amphoteric behavior of this type of nanoparticle, which allowed the nanoparticles to be more easily located on the substrate than the hydrophilic ones, preventing the surfactant from being trapped on the rock surface [43].

Even so, the mechanisms that cause the reduction in the adsorption of surfactants when using nanoparticles have not been fully elucidated either, although several theories have been raised. For example, that negatively charged nanoparticles

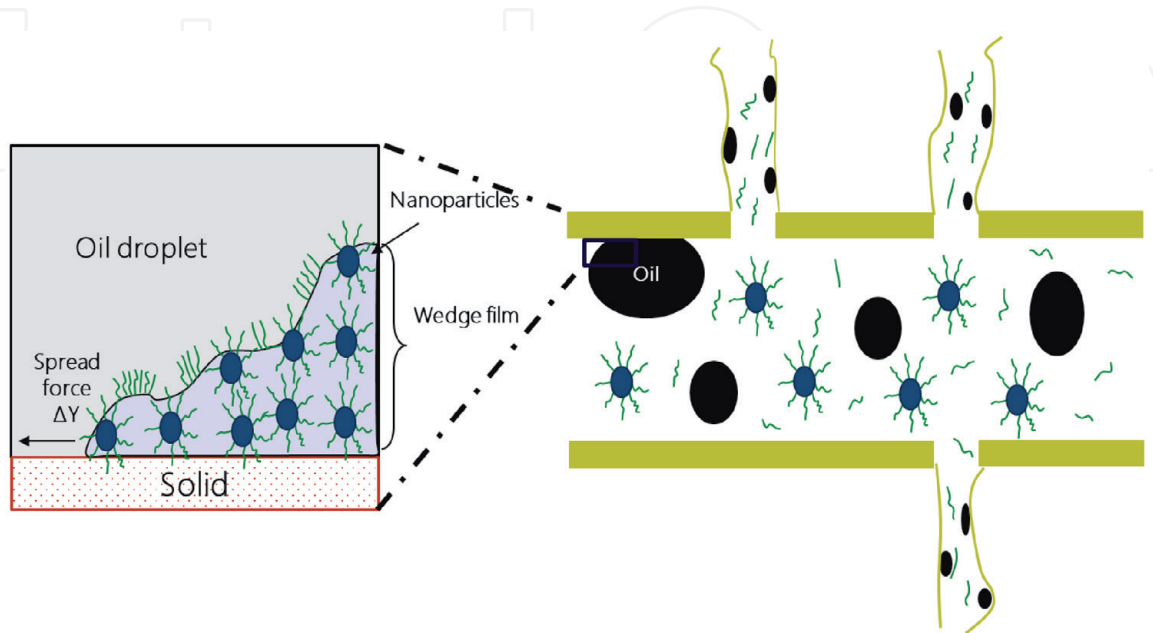


Figure 8.
Synergistic combination of surfactants and nanoparticles for EOR processes [51].

and anionic surfactant molecules compete for adsorption on the substrate surface due to electrostatic interactions of charged compounds with the solid surface [45]; for this reason, at low concentrations of surfactant this competition for being located on the surface prevents a large amount of surfactant from being located on the surface. However, by increasing the concentration of surfactant above the concentration of nanoparticles, the surfactant has a greater preference to position itself on the surface of the substrate [49]. Another theory states that the inhibition of surfactant adsorption is due to the formation of aggregates between the surfactants and the nanoparticles which tend to remain in the aqueous solution, also due to the negative charge of both the aggregates and the substrate, the repulsive forces keep the surfactants stable in the solution and prevent them from adsorbing on the surface of the substrate [50] as observed in **Figure 8**, where it is noted that the surfactant molecules adhere to the nanoparticles and prevent interactions with the substrate of the pore throats into reservoirs [51].

5. Nanoparticles influence in IFT and wettability

Additionally, of adsorption, the change of IFT and wettability are the most important EOR mechanisms reported for surfactant with nanoparticles process. Although the silica nanoparticles have demonstrated a good behavior in these topics, the use of another nanoparticle type have been more promising.

Various researches have been focused on the assisting of SDS (Sodium Dodecyl Sulfate) surfactant with other type of nanoparticle [48, 52, 53]. These studies showed that the incorporation of ZrO₂ nanoparticles allows to displace more easily, the crude trapped in the porous medium [54], since the IFT values are drastically reduced, showing the efficiency of the ZrO₂/SDS solution to reduce capillary pressure in the pore space, which allows increasing the mobilization of the crude oil [52]. Furthermore, the presence of nanoparticles can change rheological properties and increase the effectiveness of the solution in the recovery process [54].

Aluminum oxide nanoparticles also have demonstrated good performance combined with anionic surfactants displaying the effectiveness of aluminum-based nanofluids in altering the wettability of sandstone cores [55]. The results revealed that the surfactant acts by modifying the surface and that its effect can be enhanced by adding Al₂O₃ nanoparticles at low concentrations.

6. Conclusions

The surfactant injection is a promising EOR method that is in charge to reduce the interfacial tension of the water–oil interface to recover an additional amount of petroleum into the reservoir, however, a large part of the surfactant compound is entrapment on the rock surface due to interactions of the chemical and the minerals causing surfactant loss and low recovery factors.

The surfactant adsorption on the rock is a phenomenon that depends on various parameters like salinity, mineralogy, temperature, and pH which depending on the magnitude of these ones, the type of interaction of rock and surfactant will be determined. Likewise, the range of adsorption will be decided by the type of adsorption mechanism and parameters present.

The use of new techniques to reduce surfactant adsorption has been used to increase the efficiency of the recovery factor. Among the most popular, the use of nanoparticles has shown an excellent performance since reduce the adsorption of surfactant to interact with the rock surface and the surfactant as well.

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Conflict of interest

The author declares no conflict of interest.

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