Research Article

A Laboratory Approach to Measure Carbonate Rocks' Adsorption Density by Surfactant and Polymer

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Chemical recovery techniques have always been considered as one of the efficient secondary and tertiary recovery methods to enhance the oil recovery factor. Regarding the diversity of reservoir heterogeneity and rock properties for each field, various chemical agents were taken into consideration to provide a feasible process that has the best agreement with the reservoir characterization. The objective of this paper is to investigate the considerable influence of a set of chemical agents and temperature impact on the surfactant adsorption density of carbonate rocks. According to the results of this experiment, higher temperatures provide lower surfactant adsorption density. The lowest adsorption carbonate rocks' adsorption density had occurred at 80°C. Furthermore, it was witnessed that hydrolyzed polyacrylamide addition to the surfactant would cause a dramatic decrease in the adsorption density in comparison with the surfactant or polymer individually.

1. Introduction

Nowadays, many kinds of research studies have been complimented on the tertiary oil recovery field in order to enhance oil recovery (EOR) and make stable oil production after the primary and secondary oil recovery [1–7]. The use of a surfactant as a surface-active agent is one of the new techniques for growing oil extraction by changing the wettability of carbonate reservoirs from oil-wet to water-wet [8–13]. This chemical agent, by reducing IFT and/or altering surface wettability, causes the movement of trapped oils to boost and so to enhance oil recovery [14–18]. In recent decades, chemical injections have been extensively used in enhancing oil recovery, aiming at increasing the displacement coefficient in heterogeneous reservoirs and hence reducing residual oil saturation [19–24].

In petroleum engineering, the polymer is used as an additive in the petroleum reservoir to sweeping oil to increase oil recovery. Chemical flooding has subcategories such as polymer flooding, surfactant flooding, alkali/polymer flooding, and alkali/surfactant/polymer (ASP) flooding [25–31].

Injection of a surfactant and/or a polymer is one of the essential chemical injection mechanisms, especially in reservoirs with high water salinity in which alkaline is not suggested due to the deposition of salty compounds in the formation. ASP flooding can improve oil production more than water flooding noticeably; however, it causes new problems due to the attendance of high amount of alkali, for instance, the mixture of the oil-in-water emulsion has harmful effects of the shell and the corrosion of the oil production tools [32–35]. Therefore, the new surfactant

combinations without alkali are of crucial importance in the EOR process, particularly in oil reservoirs with high temperature gradient and high salinity. In this method, the surfactant can be used for two reasons: decreasing the interfacial tension between water and oil and changing rock wettability [36–39]. Additionally, using the polymer can be exploited to reduce the water and oil mobility ratio. Since a surfactant enables to bond with the strands of a polymer, the combination of surfactants and polymers creates different properties and performances than when used individually [17, 40].

Ahmed et al. (2014) evaluated the adsorption of a cationic surfactant, namely, C12 on carbonate minerals. They investigated the adsorption of this surfactant in carbonate formations and stated that its adsorption ability depends on different parameters including pH, electrolyte composition of formation water, and mineral types present in carbonate formations. The amount of its adsorption on calcite at a low level of pH (5–6.5) is $0.5 \, \text{mg/m}^2$. However, the adsorption capacity increases when carbonate formations possess silica or clay compounds. Wang et al., in 2015, worked on the adsorption of chemical surfactants on carbonate reservoirs via the injection of the surfactant/polymer in three ways: injection of the surfactant alone, injection of the polymer after the surfactant, and injection of the polymer and surfactant together. They used Betaine, an amphoteric surfactant, and hydrolyzed polyacrylamide polymer sulphonate. Results showed that the adsorption capacity of the carbonate rock was found to be 0.163 mg/g of the rock, during surfactant flooding alone, and if the injection of the surfactant along with the polymer would be fulfilled, the average capacity of adsorption was found to be 0.079 mg/g of the rock; otherwise, when the combination of the surfactant and polymer was injected, the average capacity of adsorption would be 0.083 mg/g of the rock [41]. Feng et al. in 2012 stated that the mixture of the polymer to the surfactant solution accelerated the reduction of surface tension between water and oil. They showed that the injection of chemicals containing polymer and alkaline led to increasing the oil recovery rate from 13% to 20%. This value is much larger than that when the injection of the polymer was applied alone under the same conditions [17].

Ahmed F. Belhaj et al. (2021) investigated the adsorption behavior of two chemical surfactant nanionic alkyl polyglucoside (APG) and anionic alkyl ether carboxylate (AEC) on the carbonate surfactant using static adsorption experiments and artificial neural network (ANN) prediction. The static experiment indicated that the nanionic surfactant has more adsorption density with respect to the anionic surfactant. The ANN model revealed good agreement with the experimental result, and also, the results showed that adsorption density for both surfactants decreases as temperature increases [42]. Das et al. (2020) measured the adsorption density of a nanionic surfactant with two different types of hydrophobic units and hydrophilic polyethoxylate units ranging from 15 to 40 mers on Indiana limestone. The results showed that adsorption increased by temperature increasing and decreased with more hydrophilic groups [43]. Saha et al. (2017) investigated the effect of

mineralogy on the adsorption characteristics of the surfactant-reservoir rock system. Cationic Triton X-100 was used as the chemical surfactant, and the results indicated that adsorption capacity of the surfactant is strongly dependent on the mineral content of the rock in the order of illite > feldspar > montmorillonite > kaolinite [44].

The adsorption and preservation of polymers and surfactants on reservoir rock surfaces are notable factors in the injection process due to decreasing fluid viscosity, leading to the reduction of final oil recovery. This indicates loss of chemicals from the solution and hence reduction in the amount of chemical injection. Executive and, especially, economic aspects can influence the efficiency of chemical injection operation. Therefore, the lower the absorption of the polymer and surfactant is, the lower the amount of the injectable chemical and final cost would be. Since surfactants have a high potential to reduce the surface tension between water and oil and to change the wettability of the rocks to water-wetting, the main objectives of this study are as follows:

(i) Investigation of the adsorption ability of different concentrations of Triton X-100 as a surfactant on a carbonate rock

(ii) Investigation of the impact of using the hydrolyzed acrylamide polymer on the adsorption capacity of Triton X-100 in critical concentration

(iii) Investigation of the effect of temperature on the adsorption capacity of the assessed surfactant

2. Materials and Methods

The core plug used in experiments is from the Binak oil field located in the south of Iran. The properties of the used core plug are given in Table 1.

The aqueous phase used in this study includes distilled water along with sodium chloride with the concentration of 10,000 ppm. The Triton X-100 is a nonionic surfactant with a water-wet polyethene oxide chain (5 units of ethylene oxide) and an aromatic lipophilic or hydrophobic chain and was created by Merck Company. The general properties of this material are given in Table 2.

Nonionic polyacrylamide provides a neutral solution when solved in distilled water. Table 3 and Figure 1 show the characteristics and form of this compound, respectively.

2.1. Experimental Method. The experimental procedure which was done in this investigation is as follows:

(1) Cleaning the core plug in a soxhlet by combining toluene and methanol

(2) Drying the core in the oven and powdering it with milling

(3) Prepare different concentrations of Triton X-100 (50–1000 ppm) and polymer (250–2500 ppm) in a brine solution with a concentration of 10000 ppm

(4) Separation of 3 gr of the rock powder and mixing with 60 gr of the surfactant and polymer solution

TABLE 1: The properties of the used core sample.

Material	Porosity (%)	Pore volume (cm^3)	Absolute permeability (md)	Gas permeability (md)	Diameter (cm)	Length (cm)
Anhydrite + dolomite	13.39	12.10	4.76	10.60	3.67	**8.54**

TABLE 2: The chemical and physical characteristics of the used surfactant.

Chemical formula	$C_{14}H_{22}O$ (C_2H_4O) n ($n = 9$-10)
Molecular weight	647 gr/mol
Physical state	Viscous and colorless fluid
Density	1.07 gr/cm^3
Melting point	6°C
Vapour pressure	Less than1 mmHg
Flammability point	251°C

TABLE 3: Properties of the used polymer.

Name	Partially hydrolyzed polyacrylamide
Physical state	White granule powder
Solubility	Soluble in water
Molecular weight	15–25 g/mol
Insoluble impurities	%<2.0
Hydrolization degree	20–30%
Solving time	Less than 60 min
Chemical formula	[-CH_2CH $(CONH_2)$-]
Kind	Nonionic
Free monomers and partially hydrolyzed polyacrylamide	Up to a maximum of 0.05%

(5) Perform the adsorption test in different concentrations and different temperatures

(6) Measure CMC of the surfactant solutions and polymer by electrical conductivity and pH

(7) Preparing CMC and retesting at 25, 40, 60, and 80°C

(8) Adding polyacrylamide to the surfactant solution in CMC concentration and retesting in four different temperatures and salt concentration of 10000 ppm

(9) Evaluating the results and the curves and giving recommendations

3. Results and Discussion

The adsorption of the surfactant at the salt concentration of 10000 ppm in different surfactant concentrations and four temperature ranges is primarily investigated. The critical concentration of the solutions containing Triton X-100 surfactant and hydrolyzed polyacrylamide is then calculated with pH and electrical conductivity methods, and their CMCs were used to perform adsorption tests.

3.1. Temperature Impact.
The impact of temperature on the absorption capacity of the surfactant was examined at 25, 40, 60, and 80°C. In these experiments, 3 gr of the powdered rock was placed in a beaker, and 60 g of the surfactant and brine solution with the concentration of 10000 ppm was added. Ten experiments were conducted in 24 hours with

different concentrations of the surfactant in the range of 50 to 1000 ppm. After 12 hours, the solution reached the equilibrium, and then, the adsorption density was measured using the following equation:

$$\tau = \left(\frac{(C_1 - C_e) * M_s}{M_c} \right) * 10^{-3}, \tag{1}$$

where τ is the adsorption density (mg/g), C_i is the initial concentration, C_e is the equilibrium concentration (ppm), M_s is the solution weight, and M_c is the rock powder weight (gr). The data obtained in the ambient temperature are illustrated in Figure 2. As to be seen in this graph, increasing the surfactant concentration elevates the adsorption density. The increasing rate was extremely ascending to 400 ppm and remained fairly stable. Ultimately, the adsorption density would be 3.91 mg/g at this temperature.

Previous experiments were repeated at the same concentrations at 40°C, and their results are shown in Figure 2. An increase of about 15°C has led to a decrease in density. As the data show, the concentration of the remaining surfactant in the solution has increased. The density of adsorption at this temperature has decreased to 3.556 mg/g. The experiments were conducted at 60°C, and the amount of surfactant remaining in the test container was slightly higher than that at 40°C, which means that an increase of 20°C has been able to reduce the adsorption density. The data from the experiments are shown in Figure 2. The adsorption density is reduced to 3.44 mg/g. The last tested temperature is 80°C. All tests were repeated at this temperature, and their results are shown in Figure 2. The temperature increase has a positive effect on the absorption of the surfactant. Absorption density at this temperature at the final concentration of the surfactant is 3.37 mg/g. Electrical conductivity and pH measurements were employed to obtain the critical concentration of the surfactant and polymer used in this study. In these methods, pH and electrical conductivity of different concentrations of the surfactant and polymer were measured and sketched versus concentration on a graph. The data drawn on the graph have two different trends. The trends were first regretted linearly using the linear method. Then, the critical concentration of the solutions corresponding to the intersection of the two lines was obtained. In this research, the critical concentrations of the surfactant and polymer were found to be 450 ppm and 1300 ppm, respectively.

3.2. Hydrolyzed Polyacrylamide Addition Impact.
In order to investigate the hydrolyzed polyacrylamide impact on the surfactant adsorption, 1300 ppm of this polymer was added to the surfactant solution with a concentration of 450 ppm. The obtained solution was placed on a magnetic stirrer at ambient temperature for 24 hours. It was then kept at room
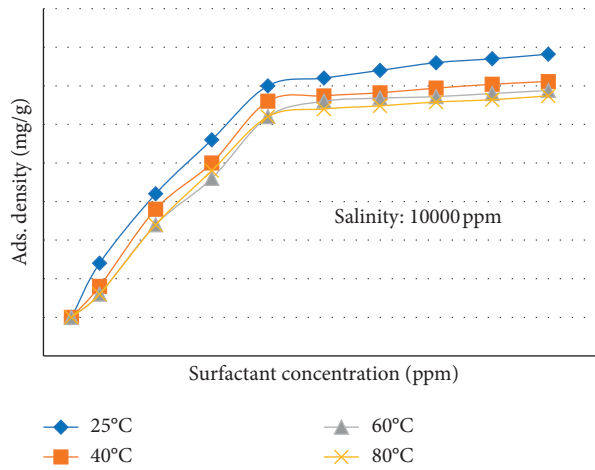
FIGURE 1: Hydrolyzed polyacrylamide.

FIGURE 2: The surfactant adsorption density vs. Triton X-100 surfactant concentration at different temperatures and salt concentration of 10000 ppm.

TABLE 4: Initial and residual values of the surfactant/polymer at ambient temperature and 40°C with a concentration of 10000 ppm.

Test no.	Used material	Initial amount of surfactant + polymer (ppm)	Residual amount of surfactant/polymer (ppm) at 25°C	Adsorption density of surfactant/polymer (mg/g) at 25°C	Residual amount of surfactant/polymer (ppm) at 40°C	Adsorption density of surfactant/polymer (mg/g) at 40°C
1	S	450	285	3.30	297.60	3.048
2	P	1300	1154	2.92	1167	2.660
3	S + P	875	757	2.36	765	2.200

S: surfactant and P: polymer.

TABLE 5: Initial and residual values of the surfactant/polymer at 60°C and 80°C with a concentration of 10000 ppm.

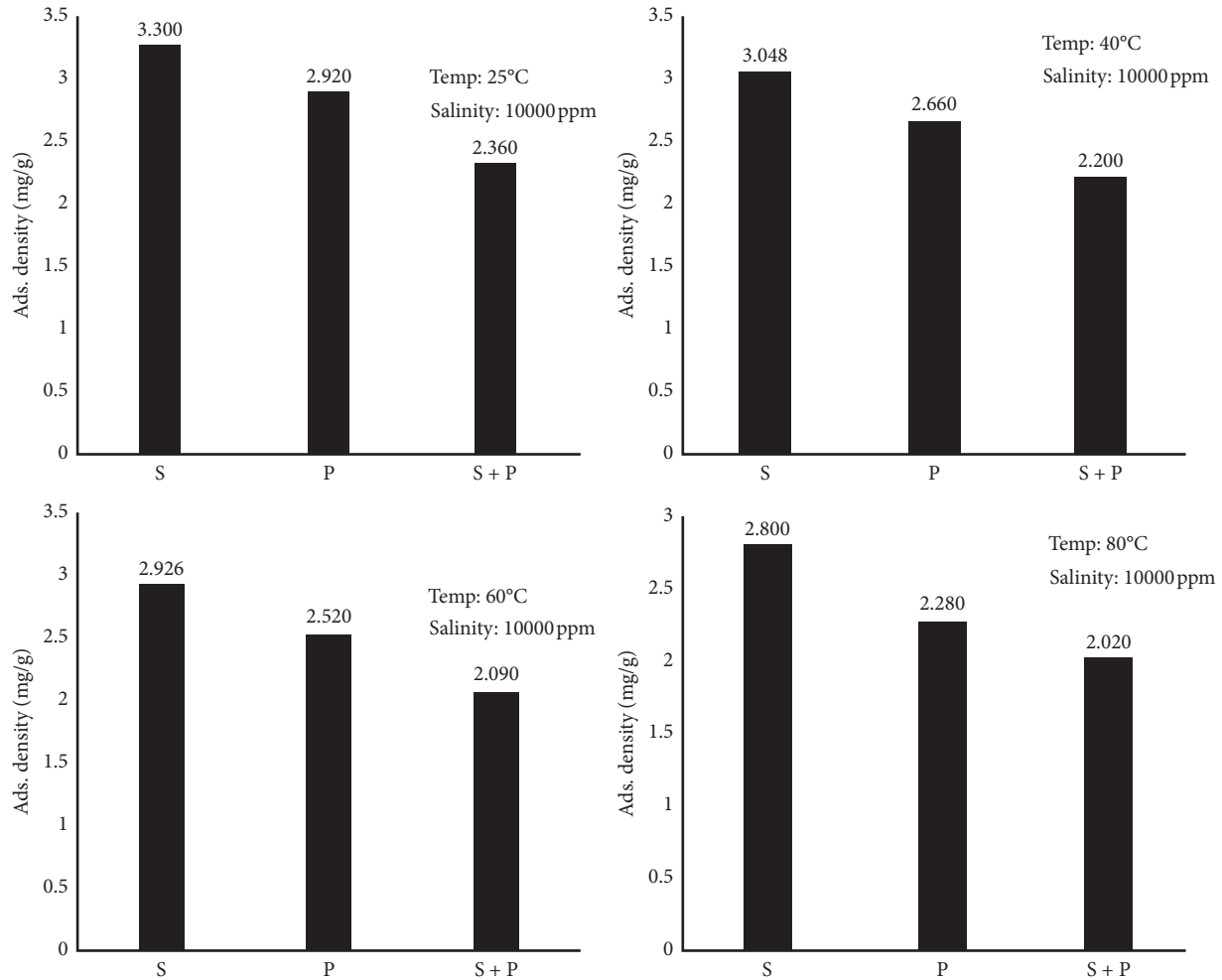| Test no.	Used material	Initial amount of surfactant + polymer (ppm)	Residual amount of surfactant/polymer (ppm) at 60°C	Adsorption density of surfactant/polymer (mg/g) at 60°C	Residual amount of surfactant/polymer (ppm) at 80°C	Adsorption density of surfactant/polymer (mg/g) at 80°C
1	S	450	303.70	2.926	310	2.80
2	P	1300	1174	2.520	1186	2.28
3	S + P	875	771	2.080	774	2.02

S: surfactant and P: polymer.

Figure 3: Polyacrylamide effect on the surfactant adsorption at different temperatures.

temperature for 12 hours to reach the equilibrium. After the equilibrium, some of the solutions were taken to measure the concentration of the surfactant by a spectrophotometer. The following tables and graphs show the results of these tests. It is concluded from the obtained data that the mixture of the polymer and surfactant could decrease the surfactant adsorption on the rock powder. Moreover, it could decrease the adsorption density from 3.3 to 2.92 mg/g at ambient temperature, 3.048 to 2.66 mg/g at 40°C, 2.926 to 2.25 mg/g at 60°C, and 2.8 to 2.28 mg/g at 80°C. The results are shown in Tables 4 and 5 as well as in Figure 3.

4. Conclusions

The main findings of this study are as follows:

  (i) The adsorption density of the surfactant or polymer reduced with increasing temperature. The maximum reduction in adsorption density was attained at 80°C for both additives.

  (ii) The polymer was prepared in 1300 ppm and added to the surfactant solution with 450 ppm. The results showed that hydrolyzed polyacrylamide had a positive effect on the reduction of the surfactant adsorption.

  (iii) The adsorption rate of Triton X-100 and/or hydrolyzed polyacrylamide alone on carbonate rock was higher than that of both compounds in a mixture of 0.027 gr surfactant and 0.078 gr polymer.

  (iv) As the temperature rises from 25°C to 80°C, the rate of adsorption of the surfactant and polymer on the carbonate rock surface decreases, and this trend is likely to continue at temperatures above 80°C; therefore, using of these compounds as a mixture (Triton X-100 and polyacrylamide) in oil reservoirs with a high temperature gradient (i.e., more than or equal to 80°C) would be suitable in terms of the economic view.

  (v) The adsorption density increases with increasing surfactant concentration up to CMC at a constant temperature, and then, it remained approximately stable.

In addition, different type probabilistic methods can be used to deal with objective uncertainties such as but not limited to [45–51].

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Acknowledgments

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