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Research Article

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Investigating Effect of SiO_2 Nanoparticle and Sodium-Dodecyl-Sulfate Surfactant on Surface Properties: Wettability Alteration and IFT Reduction

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Abstract

Interfacial-Tension (IFT)/contact angle measurements and several core-floods are performed to characterize the as-prepared samples and evaluate the efficacy of silica nanoparticles (NPs) and SDS surfactant in Enhanced-Oil-Recovery (EOR). NPs decreases the oil/solution IFT more than the surfactant solution and much more than reservoir brine. The optimum concentration of silica NPs in surfactant solution is determined to achieve in highest IFT reduction. Also, contact angle measurements unraveled the vital role of the synthesized NPs hydrophilic nature in altering the wettability to strongly water-wet. Consecutive core flood examinations corroborated the significant improvement of the surfactant performance in EOR by the NPs.

Keywords: Enhanced-Oil-Recovery (EOR); Interfacial-Tension (IFT); Wettability Alteration; Nano-Surfactant Solution; Macroscopic and Microscopic Efficiency

Introduction

The sharp increase in the ever-demanding need for safe sources of energy and the constant increment in costs have provoked many researches to innovate novel EOR methods for economical crude oil production. [1,2]. Microscopic and macroscopic efficiencies are two kinds of categories affecting the oil recovery. The efficiency coefficient of an oil reservoir, which is the multiplication of macroscopic or volumetric efficiency and microscopic efficiency, is a parameter, showing the recoverable amount of oil reserves. Hence, an increase in macroscopic or microscopic efficiencies results in higher efficiency coefficient. To increase the macroscopic efficiency, the reservoir fluid viscosity should increase and/or the mobility ratio should decrease. In addition, Interfacial-Tension (IFT) and wettability are two properties, influencing the microscopic efficiency and should be studied to plan a good flooding scenario.

One of the most common EOR approach is surfactant flooding, which has been regarded as a tertiary oil recovery. Surfactants usually have a considerable effect on IFT and wettability [3,4]. At first, Marathon Oil Company in the early 1960s presented the application of surfactants [5]. In this method, the surfactant would reduce the surface tension between water and oil phases; consequently, the capillary pressure would be reduced, and water could push the extra amount of oil [6,7]. Fu et al. presented an experimental work, which discuses about the application of a cationic surfactant for EOR purposes. They believed that using this surfactant would result in small water cut a higher oil recovery compared with before approaches [8]. A new anionic surfactant, named Ammoeng, has been used in chemical flooding, and it has been declared that it would decrease IFT between oil and surfactant solution. Furthermore, a combination solution of two surfactant is introduced, which have a great impact on IFT; finally, it has been suggested that using these surfactant solutions would be proper for enhancing the oil recovery [9]. Another liquid-base anionic surfactant was also introduced by researchers, which could tolerate in presence of high saline liquids. It was demonstrated that not only does this surfactant reduce IFT of solution and liquid phase, but also increasing in salinity would improve the IFT reduction, and it, therefore, could be used as a high-performance chemical in tertiary oil recovery [10]. It is worthy of attention that flow characteristics of a surfactant-stabilized emulsion would vary in presence of salts, and it might be problematic [11]. A research, which is done by Anganaei et al. showed that applying the coco-amido-propyl-betaine surfactant would decease IFT and enhance the oil recovery significantly due to its chemical properties [12]. Kamranfar and Jamialahmadi revealed an interesting fact that some surfactant could be used for both objectives of IFT reduction and increasing the fluid viscosity. Therefore, it holds both merits of using surfactants and polymers; as a result, oil recovery would enhance much greater than the condition when surfactants just used for IFT reduction [13]. Nowadays, nanotechnology can be considered as an influential tool in different branches of oil industry, especially in reservoir engineering. There are many attempts to explore the application of nanoparticles in EOR [14]. In 2006, the performance of nanoparticles to alter the porous media wettability was studied, and it was concluded that the oil recovery would be improved by using some nanoparticles, especially in concentrations ranging from 0.02 to 0.03 [15]. Then, it was found that nanofluids, considerably, would improve oil recovery by reduction in IFT [16]. Nanoparticles along with surfactants would reduce IFT and surfactant adsorption amount; as a consequence, it would enhance the oil recovery sharply in comparison with surfactant flooding alone [14,17]. Additionally, using nano-surfactant solutions for the chemical flooding scenarios would create a strong hydrogen bond between silica and water; so, surface energy would increase. On this account, Medium wettability would alter from petrophilic to hydrophilic [1]. As a result, nano-surfactant solutions should to be considered as a high potentially method to enhance the oil recovery [18].

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Recently, it has been stated that in nano-EOR approaches, IFT plays a much more important role to enhance the oil recovery in comparison with wettability values. Moreover, researchers performed some core flooding tests and revealed that silica nanofluids could be used for a wide range of reservoir wettability at the reservoir temperature [19]. The effect of hydrophilic silica nanoparticles was studied on the amount of recoverable oil through a silica bead column. It has been reported that these nanoparticles would assist the recovery process since they increase the mobility of oil in the porous media [20].

In this experimental research, SiO_2 nanoparticles are used along with SDS, as an anionic surfactant, to explore the extent of these chemical effects on IFT, wettability, and amount of recoverable oil as a result of several bench test and sandstone core floods. The SiO_2 nanoparticles were characterized by Transmission-Electron-Microscopy (TEM), X-Ray Diffraction (XRD), and Fourier-Transform Infrared Spectroscopy (FT-IR). To have more reliable results, first, IFT values of reservoir oil with diluted reservoir brine, surfactant solution, and several nano-surfactant solutions with different nanoparticle concentrations were determined; thereafter, for these conditions, contact angles were measured to investigate the effect of nanoparticles and surfactant on the reservoir rock wettability. Finally, three core flood tests were conducted to observe the different chemicals effects on the oil recovery through the porous media at ambient conditions.

Materials and Methodology

SDS surfactant

SDS is bought from a reliable and expert local company (SinaClon). CMC value between the surfactant and diluted reservoir brine (2000 ppm) was approximately 2150 ppm, determined by conductivity measurement technique.

Solution characterization

XRD patterns of the as-prepared samples were obtained and collected 2θ data from 0 to 100° by Cu Kα radiation (λ=1.5404) in a Philips PW 1142-Netherland to identify the final phase of the samples. TEM images were captured using Zeiss-EM10C-80 KV. FT-IR spectroscopy were collected in 400-4000 Cm^{-1} range by Bruker Optics TENSOR 27 spectrometer at ambient condition. XRD pattern of SiO_2 nanoparticles is shown in Figure 1. According to Debye-Scherrer formula, $t = 0.9\lambda/\beta cos\theta$, the maximum average size of particles (25 nm) was calculated.

TEM images of SiO_2 nanoparticles are shown in Figure 2. It can be seen that the morphology and particle size of synthesized samples is in order of 20-30 nm, which is well-consistent with XRD in Figure 1.

FT-IR spectrum of SiO_2 nanoparticles, which is in the range of 400-4000 cm^{-1}, can be observed in Figure 3. The strong peak at around 1106 cm^{-1}, as a stretching vibration, can be ascribed to Si-O bond, which is reported IR spectra for SiO_2 in the literature [21]. Furthermore, a broad peak around 2900-3500 cm^{-1} can be assigned as stretching vibrations of amino groups [22].

Oil, brine, and core samples

Reservoir oil and brine and a sandstone core plug, which the flooding tests would be done through it, were sampled from an Iranian reservoir. Properties of crude oil and sandstone core sample are mentioned in Table 1. The brine, used in this study, is diluted reservoir brine with concentration of 2000 ppm.

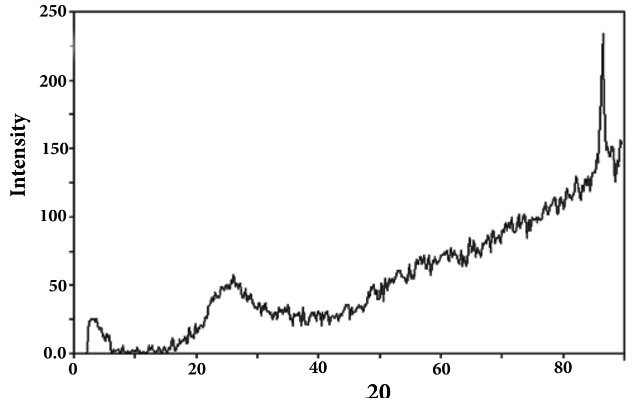

Figure 1: XRD spectrum of SiO_2 nanoparticles.

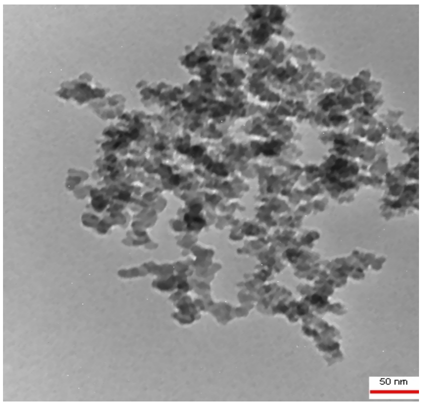

Figure 2: TEM image of SiO_2 nanoparticles.

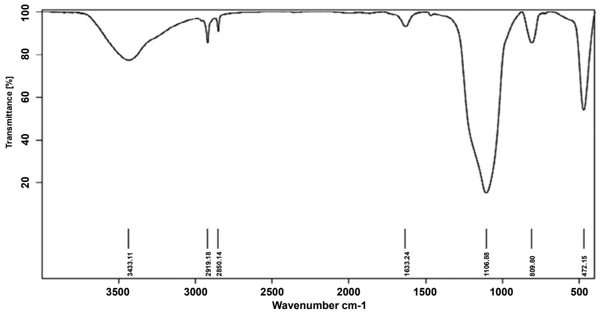

Figure 3: FT-IR spectroscopy of SiO_2 nanoparticle.

Reservoir oil	Properties	
	Viscosity (cp)	˚API
	45	37

Sandstone core sample	Properties						
	Porosity (fraction)	Permeability (md)	Length (cm)	Diameter (cm)	Pore volume (cc)	Water saturation (fraction)	Oil saturation (fraction)
	0.2	200	14	3.8	31.7	0.11	0.89

Table 1: Physical and chemical properties of crude oil and sandstone core sample.

Solution preparation

To prepare the surfactant solution, surfactant was dissolved in the brine using the magnetic stirrer for 3 hours. Afterwards, the mixture was equilibrated for 24 hours, and, then, it was centrifuged for 15 minutes at 2000 rpm. After the preparation of this solution, to prepare the nano-surfactant solution, nanoparticles were added to it and stirred. Then, nanoparticles were dispersed in the solution using the ultrasonic device. After that, to check the stability of the prepared solution, it was maintained in a closed transparent glass container, away from the destructive agents such as Ultraviolet (UV) light and heat for about one week. Finally, five solutions were prepared with constant concentration of SDS surfactant (2150 ppm) and five different SiO_2 nanoparticles concentrations with the weight percent of 0, 0.05, 0.10, 0.15, and 0.20.

IFT measurements

IFT determinations were performed to study the surfactant and nanoparticle effects and to select the optimum nanoparticle concentration for nano-surfactant solutions. IFT values between different aqueous solutions of diluted reservoir brine, surfactant, and nano-surfactant with crude oil were measured using the pendant-drop technique at ambient conditions (pressure of 14.7 psi and temperature of 30°C). In addition to the brine and surfactant solution, the best nano-surfactant solution, which has the lowest IFT in adjacency with reservoir oil, has been selected. Schematic view of pendant-drop apparatus is shown in Figure 4. The key point about IFT measurements is using optimal experiment conditions. More precisely, in a pendant drop setup, different needles could be used according to the tests fluid and their fluid properties. For instance, in a liquid-liquid system of brine and light oil, which is the subject of this work, the needle diameter should be 1.5 mm. However, if heavy oil be in the system, its diameter is better to change to 0.69 mm to have optimal conditions and to obtain reliable results.

Wettability measurements

To examine the effect of surfactant and nanoparticles on the porous medium wettability, sessile drop method was applied. Utilization of surfactant reduces interfacial tension between fluids in porous medium and also causes wettability alteration. Nanoparticles are also used for wettability alteration in porous media. Wettability alteration effect by mixing the effect of interfacial tension reduction because of nanoparticles and surfactant is considered due to surface chemical properties and reaction of fluid and rock surface. Schematic view of this setup is shown in Figure 5. To perform a test, small slices of a clean core plug were cut and polished. The slices were aged with the crude oil at high pressure and temperature for two weeks. Then, the slices were washed with *n*-heptane; finally, they were dried in an oven for about 12 hours. Next, slices were submerged in three containers of the reservoir brine, surfactant solution, and nano-surfactant solution for about 7 days at ambient condition. Then, the contact angles were measured between oil phase and three solutions of diluted reservoir brine, surfactant solution, and nano-surfactant solution.

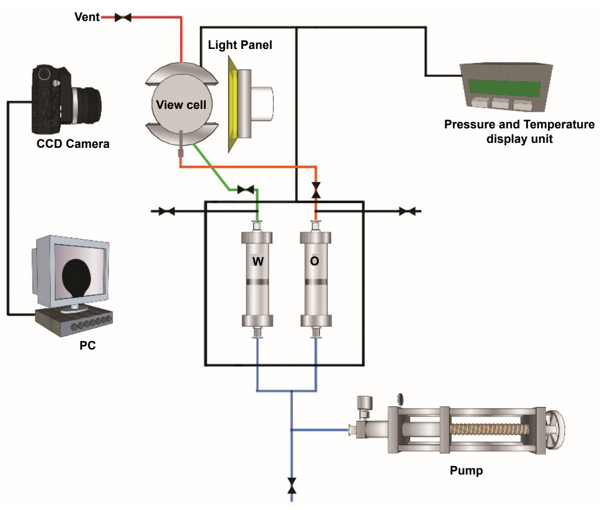

Figure 4: Schematic of interfacial tension measurements apparatus.

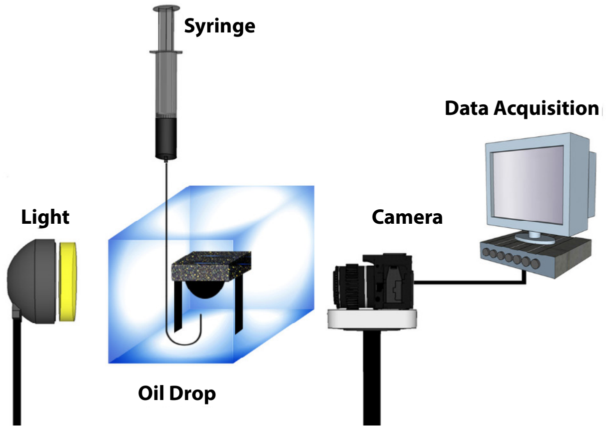

Figure 5: Schematic of contact angle measurement apparatus.

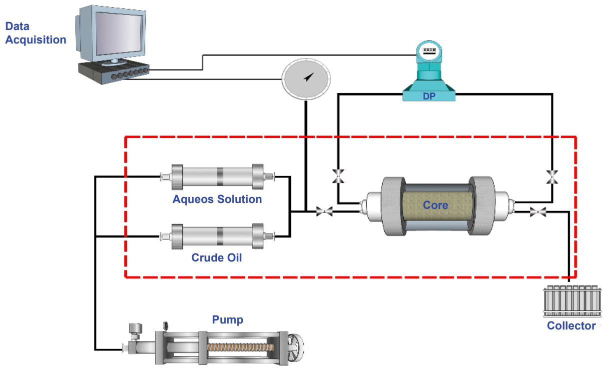

Figure 6: Schematic view of core flooding system.

Flooding tests

Nanoparticles and surfactants improve microscopic displacement efficiency and increase oil recovery from porous medium by two principal mechanisms, reducing interfacial tension and wettability alteration. Therefore, residual oil saturation decreases. Core flooding apparatus, which is shown in Figure 6, consists of a core holder, a pump with a constant injection rate, transfer vessels, pressure transducers, and data acquisition system. Prior to performing experiments, the core sample was cleaned and vacuumed. To run a test, the core sample was placed in the core holder and was connected to the system by inlet and outlet steel pipes. Afterwards, it was saturated with diluted reservoir brine. To establish irreducible water saturation, crude oil was injected at a constant flow rate until no water produced. Injections of brine, surfactant, and nano-surfactant solutions into the core sample was done with a constant flow rate of 20 cc/hr.

Obtained results and analysis of experimental results

According to Table 2, which shows the IFT results, it has been observed that the oil/brine IFT is 32 dyne/cm. Oil/surfactant solution has an IFT of 18.2 dyne/cm, and presence of nanoparticles in surfactant solution leads to more reduction of IFT to a value of 4.7 dyne/cm. The associated uncertainty with IFT measurements is ±0.5 dyne/cm, which demonstrates that the measured IFT values are reliable, and difference among the measured IFT values is not appeared from the uncertainty of measurements and is happened as a result of different nanoparticles concentrations. Interfacial tension decreases by increasing surfactant concentration up to CMC critical concentration, then remains stable and have not tangible change. Solution containing both nanoparticles and surfactant, reduces interfacial tension up to critical concentration more than before, and a slight increase in interfacial tension is observed after this concentration. Therefore, the optimum concentration of SiO_2 nanoparticles is 0.1 wt%, which resulted in the lowest IFT. Moreover, this nanoparticle concentration has also economic justification. Regarding the literatures, presence of nanoparticles reduces IFT due to the formation a mixed layer with surfactant at the interface of two phases [18]. As the nanoparticle concentration increases more than a certain value, IFT of oil/nano-surfactant solution would slightly increase due to the high concentration of nanoparticles at their interface, which do not allow surfactant molecules to place at the interface [17].

Experimental phases			IFT (dynes/cm)
Heavy phase		Light phase	
Reservoir brine (2000 ppm)		Reservoir oil	32
Optimum surfactant solution (CMC=2150 ppm)		Reservoir oil	18.2
Nano-surfactant solution (CMC : 2150 ppm)	1-4: nanoparticle concentration (0.05 wt. %)	Reservoir oil	5.8
	2-4: nanoparticle concentration (0.1 wt. %)	Reservoir oil	4.7
	3-4: nanoparticle concentration (0.15 wt. %)	Reservoir oil	5.4
	4-4: nanoparticle concentration (0.2 wt. %)	Reservoir oil	6.2

Table 2: Liquid-liquid IFT measurements for oil/aqueous solutions systems.

Type of specimen	Degrees at four points			
	1	2	3	4
Influenced tablet by nano-surfactant solution	21	20.8	20.2	21.5
Influenced tablet by surfactant solution	26.2	25	24.3	29.6
Influenced tablet by reservoir brine	140	151	145	149

Table 3: Contact angles for different solutions using sessile drop method.

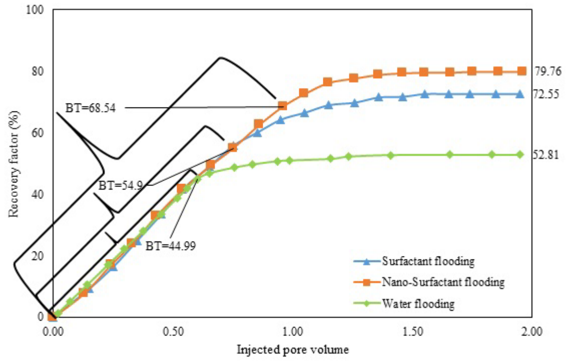

Figure 7: Oil recovery versus injected pore volume for water flooding, surfactant flooding, and nano-surfactant flooding at reservoir salinity of 140000 ppm and temperature of 30˚C (including ultimate oil recovery and breakthrough time (BT).

Table 3 presents measured contact angles in various conditions. Wettability alteration due to surfactants and existence of nanoparticles have done experimentally before. Successful application of this method strongly depends on target reservoir conditions. Wettability tests have been performed using reservoir brine, surfactant solution, and nano-surfactant solution at the optimum nanoparticle concentration, demonstrated by IFT measurements. Based on these data sets, the average contact angle of reservoir brine and oil was 146°. Therefore, the porous media was oil-wet and the aging process has been done successfully. The average surfactant solution contact angle and oil was 26°. It is a sign of water-wet system as a result of surfactant solution. Furthermore, silica nanoparticles could reduce the contact angle to a value of 21° and alter the porous media wettability to the strongly water-wet. These results demonstrate that silica nanoparticles with their hydrophilic nature and their adsorption onto the rock surface (, which is inspired by contact angle measurements) can change the wettability to strongly water-wet.

Based on IFT and contact angle data, three core-floods, including water flooding, surfactant flooding, and the optimum nano-surfactant solution flooding were performed on the sandstone core sample. Figure 7 shows the oil recovery during two injected pore volumes. Obtained data shows that nano-surfactant solution has the highest oil recovery. The oil recovery for water flooding, surfactant flooding, and optimum nano-solution flooding was around 53%, 73%, and 80%. As it could be seen from these recoveries, nano-surfactant solution dramatically increases the oil recovery. Adding the nanoparticles to the surfactant solution leads to delay in injected solution Breakthrough-Time (BT). BT is the period between the beginning of injection and appearance of injection fluid at the outlet of the porous media. Longer BT is the result of improved injected fluid and reservoir oil mobility ratio. Based on the literatures, it is supposed that modification of nano-surfactant rheological properties is due to its non-Newtonian flow characteristic [18], which leads to an increase in the viscosity of injected fluid; according to observation and laboratory measurements, which are done using a Thermo-Haake VT550 viscometer using standard NV and SV1 cup-and-spindle sensors, viscosities of diluted reservoir brine, surfactant solution, and optimum nano-surfactant solutions are 1.02 cp, 1.26 cp, and 5.2 cp, respectively. Using surfactant and nanoparticles increases injection fluid breakthrough time and also oil recovery. Moreover, in nanofluid and surfactant flooding, oil recovery increases after breakthrough time. Thus, silica nanoparticles could efficiently enhance the surfactant performance by improving microscopic and macroscopic efficiencies, and, consequently, oil production and recovery factor also increase. In macroscopic scale, increasing the injected fluid viscosity leads to higher oil recovery (mostly before BT), and in microscopic scale, IFT reduction and wettability alteration from oil-wet to water-wet condition would enhance the oil recovery (mostly after BT), both of which are resulted in differences among the three curves of Figure 7.

Conclusions

A series of tests, namely IFT measurement, contact angle measurement, and core-floods, have been conducted to select the best EOR method in the presence of nanoparticles and surfactant. Relying on the obtained results, following points can be concluded:

- According to the IFT measurements, using SDS anionic surfactants reduces the IFT between oil and aqueous solution. In addition, dispersing the silica nanoparticles lead to more reduction in the IFT values. Moreover, the nano-surfactant solution could alter the reservoir rock wettability to strongly water-wet.

- An optimum nanoparticle concentration should be selected before the flooding tests. Hence, oil recovery would be maximum if the optimum solution injected through the porous media.

- Breakthrough of the injection fluid might be occurred later in the presence of nano-surfactant solution since the reduction of mobility ratio would increase the macroscopic efficiency.

- IFT and wettability roles are bolder after the breakthrough time. To clarify, they would increase the microscopic efficiency noticeably in comparison to mobility ratio.

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