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STUDY ON DIFFERENT CONCENTRATIONS OF NANOPARTICLES AS AGENT OF OIL RECOVERY EFFICIENCY IN HIGH SALINITY RESERVOIR

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Abstract

Most oil reservoirs around the world are experiencing their second half of life. Hence, the necessity of appropriate enhanced oil recovery (EOR) method as a more efficient technology gets further importance. Nanotechnology is an advanced technology that has proved its potential to enhance oil recovery. Innovative techniques are highly required in the oil and gas industry to meet the future demand of the hydrocarbon fuels; significant efforts have been exerted on the development of new innovative technologies to increase the oil recovery. Nanotechnology has provided new promising approaches to enhanced oil recovery (EOR) techniques in matured and marginal fields. In this research, the different concentrations of TiO_2 and SiO_2 nanoparticles were formulated and their properties have been measured in terms of their application in enhancing oil recovery efficiency in high salinity reservoir conditions. It was observed that 3wt% SiO_2 nanoparticles solution yielded the highest tertiary recovery of 26.67% compared to the 0.01wt% TiO_2 nanoparticles solution that yielded 23.33%. SiO_2 Nanoparticles solutions also showed significant reduction in IFT values and contact angle measurements compared to TiO_2 nanoparticle solutions. 3wt% SiO_2 concentration had the lowest IFT value of 5.26 dyne/cm and lowest contact angle of 12.09 ° compared to all the different nanoparticle concentrations used in this research. Thus, it was concluded that 3wt% SiO_2 nanoparticles solution was the optimum concentration to be used in high salinity reservoir condition.

Keywords: Titanium dioxide nanoparticles (TiO_2); Silicon dioxide nanoparticles (SiO_2); Brine (NaCl); Sand pack; IFT; contact angle measurement.

1. Introduction

The current growing demand for oil led by major energy consuming countries has become a critical challenge for the oil industry to secure new oil resources. As a result, many companies all over the world are investing in enhanced oil recovery projects for their producing reservoirs. Nanotechnology is the use of very small pieces of material, at dimensions between approximately 1-100 nanometers by themselves or their manipulation to create new large-scale materials, where unique phenomena enable novel applications. The application of nanoparticles (NP) has been applied in many areas such as: electrical Industry, mechanical industry, oil and gas industry [1-4]. Nanotechnology is applied in the oil and gas industries in areas such as enhancing oil recovery (Nano-EOR), improving the equipment performance, reducing energy losses during production, providing real-time analysis on emulsion characteristics by using nanoparticles, nano-fluids or nano-emulsions.

The performances of nanoparticles in EOR differ from one another [5]. Such nanoparticles include titanium oxide, tin oxide, iron oxide, magnesium oxide, zinc oxide and many others. Nanoparticles are proven to possess physical properties such as thermal conductivity, thermal diffusivity and connective heat transfer coefficient compared to those of base fluid like oil and water [6]. The main aim of using nanoparticles in EOR is to increase oil recovery. However, the application of nanoparticle injections into the reservoir is governed by certain parameters such as concentration of nanoparticles, size, salinity, reservoir temperature, injection rate, rock grain size, clay content and reservoir permeability. If these parameters are not controlled, oil recovery will not be increased [7]. Recent studies have proven that 15% of the residual oil that is trapped in the reservoir can be recovered by nanoparticles [8]. The residual oil is recovered from the reservoir by mixing the nanoparticles in water to create a river-like flow called Nano scale traffic jam [9]. It was reported that as the particle propagates through the porous media, several transport mechanisms result in a decrease in the dispersed silica concentration. The main limiting cause of the retention of nanoparticles in porous media is the adhesion on the pore walls and blocking of the pore throats [10-12]. Blocking of the pore throats can be caused by two mechanisms namely mechanical entrapment and log jamming (accumulation) [13]. The nanoparticles block the unwanted path away for oil (pores media tunnel) and forces the oil to move through another path, which will accumulate the oil in production area to increase the production rate. This mechanism could happen due to the size of the nanoparticles that can move faster than oil or water particle; therefore, it can block the pores tunnel. The ability of nanoparticles to alter the wettability and reduce the interfacial tension between the oil and water has been actively investigated [14]. Several authors have published the use of nanoparticles to alter wettability and its following effect on oil recovery [15-26]. The focus of the investigation is on the nanoparticle particles such as applications in high temperature and high-pressure reservoir, and also in high salinity reservoir, it also has been proven that nanoparticles are capable of recovering heavy crude oil such as asphaltenes and paraffin that contain crude oil [27].

In this research, the study will be conducted by analyzing the concentration of the nanoparticles when injected into the reservoir and how this parameter can affect the oil recovery. The nanoparticles used in this research are mainly silicon dioxide and titanium dioxide nanoparticles.

2. Materials and methodology

2.1. Materials

Table 1. Properties of Nanoparticles

Properties	TiO_2	SiO_2
Appearance	White nanopowder	Dispersion
Particle size	<25 nm	<25 nm
Surface area	44 - 55m^2/g	75.5 m^2/g
Assay	99.7 % trace metal basis	99.99% trace metal basis
Density	3.9 g/mL at 25℃	1.087 g/mL at 25℃

The core samples were obtained from CORE laboratory Sdn. Bhd in Shah Alam, Malaysia. Silicon dioxide and titanium dioxide nanoparticles were used for this experiment. Brine with concentration of 7wt.% was used in the formulation of the nanoparticle solutions. The sand sample used for the sand packing analysis was collected from Port Dickson Beach, Malaysia. The characterristics of the nanoparticles are shown in Table 1.

2.2. Methodology

The experiment consisted of using different concentration of nanoparticles to investigate its effect in enhancing oil recovery in a high salinity reservoir condition. The proposed concentrations chosen were 0.01 wt.% TiO_2, 0.1 wt.% TiO_2, 0.5 wt.% TiO_2, 0.03 wt.% SiO_2, 0.3 wt.% SiO_2 and 3 wt.% SiO_2. The optimum concentration of the nanoparticles were selected based on the results and analysis drawn from the interfacial tension measurement, contact angle measurement and performance of nanoparticles in the sand packing experiment.

The experiment conducted for this research were basically divided into three main categories, namely fluid measurement, contact angle measurement, and sand pack analysis. In the fluid measurement, the API gravity of the crude oil to be used in the experiment was measured to determine its suitability in subsequent experiment. This was then followed by the formulation of solutions. The required amounts of different nanoparticles to be used were measured and diluted with distilled water up to 200mL. The densities of the nanoparticle solutions were then measured for viscosity calculation and also to evaluate if the addition of the nanoparticles could increase the oil recovery. The densities were conducted by measuring the weight of the fluid in a known volume. Lastly, interfacial tension (IFT) measurement was conducted between the crude oil and the formulated nanoparticle solutions to evaluate how the addition of nanoparticle solutions at different concentrations could improve the oil recovery. Du Nuoy ring method was used for the IFT measurement.

Before the contact angle measurement, the sandstone cores were saturated using different concentrations of TiO_2 Nano fluids, different concentrations of SiO_2 Nano fluids and brine. Core saturation was done to evaluate the effect of each fluid on the properties of the cores. Contact angle measurement was preceded after the fluid measurements were completed. The contact angle test was performed mainly to evaluate if the addition of nanoparticles could improve the wettability of the core by changing fluid (crude oil) deposited on the core from oil-wet to water-wet. If the wettability is changed from oil wet to water-wet, oil recovery is improved. Contact angle measurement was done using contact angle analyzer- the Phoenix series.

Sand pack analysis was the final experiment of the research. Prior to the sand pack experiment, the porosity and permeability of the sand sample were measured. The porosity of the sand sample was determined by using a direct method where the bulk volume of a consolidated porous sample was determined and then the volume of the material was also found while permeability of the sand sample was determined by using constant head method to measure the coefficient of permeability (K) and then relating the coefficient of permeability (K) to the Darcy equation to determine the permeability of the sand sample. Sieving analysis was also carried out on the sand sample so as to determine its characteristics and grain distribution and also to determine if the sand sample will be suitable for use in the sand packing experiment. Once the grain distribution of the sand sample was confirmed, the sand was then built into a column to simulate a reservoir and then saturated with crude oil. Brine solution was used to simulate secondary recovery while nanoparticle solutions were used to simulate tertiary recovery. Sand pack experiment was conducted to assess how the addition of nanoparticles at different concentrations could enhance tertiary recovery in a high reservoir salinity condition.

3. Results and discussion

Fluid measurement

Characteristics of crude oil

The API gravity, viscosity and density of the crude oil was determined. The results of the measurements are shown in Table 2.

Table 2. Properties of crude oil

Property	Value
Viscosity	1.1772 cP
API gravity	32.40
Density	0.8609 g/mL

By comparing the result of the ° API gravity obtained in the experiment to the standard API classification of crude oil shown in Table 2, it can be seen clearly that the crude oil used in this experiment, falls in the 'light oil' category with an °API of 32.40.

IFT measurement

Interfacial tension relates to the capillary number, which is one of the main principles controlled in enhanced oil recovery. By lowering the IFT, it will result in a lower capillary force and thus a higher capillary number that is very desirable in enhanced oil recovery. Lower IFT results means less retention of oil, which would result in lower residual oil saturation. The IFT between the crude oil and nano-particles solutions were measured using the Du Nuoy ring technique. The results of the measurement were summarized in the Table 3 and Fig. 1.

Table 3. Results of IFT measurement of crude oil and NP solution

Solution	IFT (dyne/cm)
Base solution (Brine solution with crude oil)	11.83946
NP solution 1 (0.01wt% TiO_2)	5.41801
NP solution 2 (0.1wt% TiO_2)	6.28518
NP solution 3 (0.5wt% TiO_2)	6.71892
NP solution 4 (0.03wt% SiO_2)	6.62938
NP solution 5 (0.3wt% SiO_2)	5.48645
NP solution 6 (3wt% SiO_2)	5.25831

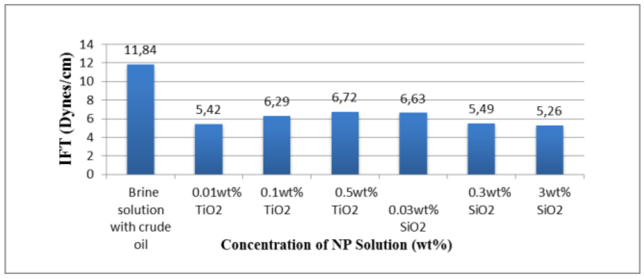

Figure 1. Graphical representation of IFT measurement

The interfacial tension between the crude oil and nanoparticle solutions were measured in dynes/cm which is equivalent to mN/M. Initially, the base solution which consisted of only brine solution was used to measure the interfacial tension. From the result, the IFT value obtained using only brine solution was 11.84 dynes/cm. However, when the nanoparticle solutions were introduced during measurement, the IFT values reduced significantly to 5.26 dynes/cm. This could be seen in the graphical representation of the IFT measurement shown in Fig. 1.

From the graph, the lowest concentration of TiO_2 nanoparticles (0.01 wt.%) yielded a low IFT value while the highest concentration of SiO_2 nanoparticle (3 wt.%) yielded the lowest IFT value. By comparing the concentrations of TiO_2 and SiO_2 nanoparticles used in the experiment, it indicates that the SiO_2 nanoparticles with 3wt % concentration had the lowest IFT value of 5.26 dynes/cm compared to TiO_2 nanoparticles with 0.01 wt.% concentration which yielded a low IFT value of 5.43 dynes/cm. Thus, it can be concluded that this concentration is considered as the optimum concentration that could be used to improve enhanced oil recovery. From the results obtained, it can be inferred that SiO_2 nanoparticles are more suitable in enhancing oil recovery in high salinity reservoir compared to TiO_2 nanoparticles.

Wettability measurement

Table 4. Result of contact angle measurement

Solution	Contact angle
Crude oil before core saturation	36.77°
0.01 wt.% TiO_2 after core saturation	13.90°
0.1 wt.% TiO_2 after core saturation	18.49°
0.5 wt.% TiO_2 after core saturation	20.36°
0.03 wt.% SiO_2 after core saturation	15.92°
0.3 wt.% SiO_2 after core saturation	12.87°
3 wt.% SiO_2 after core saturation	12.09°

Wettability is defined by the contact angle of the fluid with the solid phase. In this analysis, contact angle test was conducted to analyze how the addition of nanoparticles of different concentration could improve the wettability and also to determine the optimum concentration that is suitable to enhance oil recovery. In this case, the contact was analyzed between the sandstone cores and crude oil. Prior to the contact angle test, sandstone core were soaked in different concentrations of NP solutions and the contact angle before core saturation was also measured. The results of the analysis are shown in the Table 4.

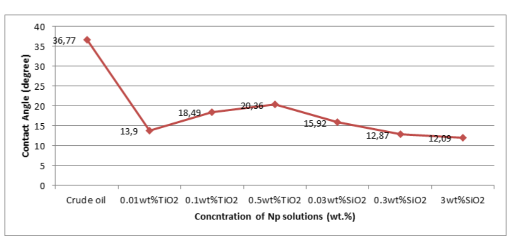

Figure 2. Graphical representation of contact angle measurement

From the graph shown above, it can be seen that all the concentrations used in this experiment yielded low contact angles compared to the initial contact angle measured before core saturation with nanoparticles.

By comparing the concentration used, it was seen clearly that SiO_2 NP solutions had a lower contact angle to TiO_2 NP solutions. The SiO_2 NP solution with concentration 3 wt.% had the lowest contact angle (12.09°) compared to all the concentrations used in this experiment. From this analysis it can be inferred that SiO_2 nanoparticles are more suitable to be used to enhance oil recovery in high salinity reservoir condition. It can also be deduced that the optimum concentration and best nanoparticle solution that can be used is the one with the lowest contact angle that is able to move the residual oil easily in from the reservoir to the surface. In this case, the optimum concentration that can be used to enhance oil recovery is 3wt% SiO_2 NP solution.

In order to validate the optimum concentration that was found from the wettability analysis, sand packing experiment was then conducted using all the different concentrations of the nanoparticle solutions.

Sand packing analysis

This is one of the alternative methods used to determine oil recovery [10]. In this method, sand was built into a column to simulate a reservoir and then saturated with crude oil. Brine solution was used to simulate secondary recovery while nanoparticle solutions were used to simulate tertiary recovery. Sand pack experiment was conducted to determine the performance of the different concentration of the nanoparticles in enhancing oil recovery from a high salinity reservoir conditions and also to determine the optimum concentration and nanoparticle that is suitable for a high salinity reservoir condition. A high salinity reservoir condition for the sand packing was achieved by injecting high saline water of about 70000 ppm into the sand pack and allowing the saline water to saturate in the sand column. The results of the experiment are shown in Figs. 3 and 4.

By comparing the different concentrations of nanoparticle solutions used with respect to the oil recovery, it was found that the 3 wt.% SiO_2 NP yielded the highest tertiary recovery of 26.67% compared to the 0.01wt% TiO2 NP solution that yield 23.33%. The other concentration used also produced tertiary recovery but their yield were not much as compared to the 3wt% SiO_2 NP and 0.01wt% TiO_2 NP solution. It was observed that in the TiO_2 NP solution, the higher the concentration of the nanoparticle solution, the lower the yield of the tertiary recovery. In the case of the SiO_2 nanoparticle solutions, it was observed that the higher the concentration of the nanoparticle solution, the higher the yield of the oil recovery and vice versa. This result shows that the type of nanoparticles used in EOR has a significant impact on how much oil can be recovered. However, at all concentrations, both SiO^2 and TiO^2 nanoparticles show improvement in oil recovery.

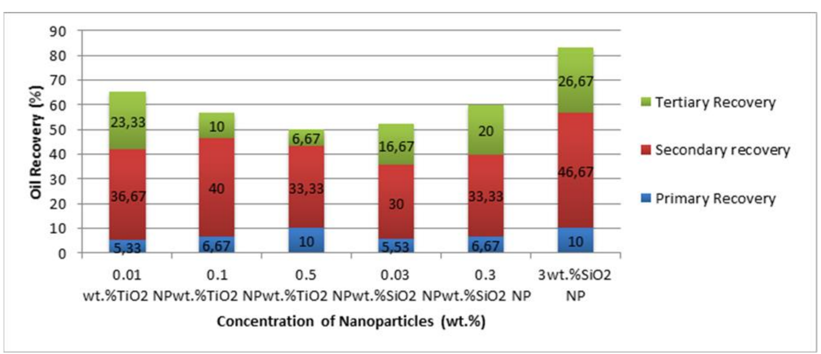

Figure 3. Graphical representation of oil recovery versus concentration of NP

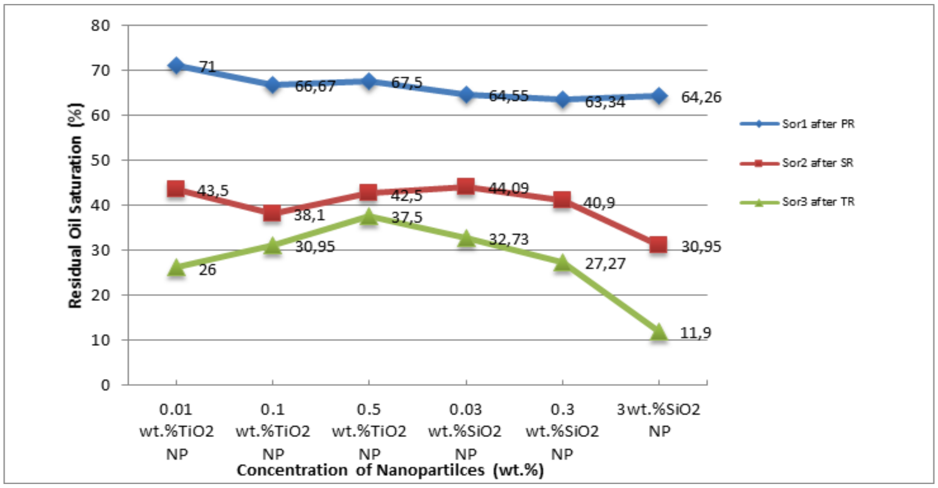

Figure 4. Graphical representation of residual oil saturation versus concentration

In terms of the residual oil saturation, the addition of nanoparticles in the tertiary recovery reduced the residual oil saturation significantly as compared to both the primary and secondary recovery. SiO_2 nanoparticles with concentration with 3wt.% had the lowest residual oil saturation with a value of 11.9% followed by 0.01 wt.% TiO_2 nanoparticles that had a residual oil saturation of 26%. From this analysis, it can be stated clearly that the SiO_2 NP solutions surpassed the TiO_2 NP solution in enhancing oil recovery in a high salinity reservoir condition. Hence it can be deduced that SiO_2 NP solutions are the most suitable type of nanoparticle to be used in high salinity reservoir condition and from the concentrations that were used in this research, the optimum concentration and nanoparticle solution that can be used is the SiO_2 NP with a concentration of 3wt%.

4. Conclusion

For this research, the different concentrations of TiO_2 and SiO_2 NP were formulated and their properties have been measured in terms of their application in enhancing oil recovery. Among the different concentration of NP solution used, it was found that 3wt% SiO_2 NP solution was the optimum concentration to be used in a high salinity reservoir condition. This concentration yielded the lowest values for both IFT and contact angle measurements. Also in terms of the sand pack analysis, it was discovered that 3wt% SiO_2 NP solution yielded the highest tertiary recovery with the lowest residual oil saturation compared to various concentrations of TiO_2 NP solutions. From the experiment conducted, the optimum concentration that can be used to increase the efficiency of oil recovery in a high salinity reservoir condition is 3wt% SiO_2 NP solution. Finally, it can be concluded that the nanoparticles contributed greatly in the IFT reduction and wettability alteration, which improved the oil recovery significantly.

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