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Effects of particle shape and size on nanofluid properties for potential Enhanced Oil Recovery (EOR)

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Abstract. Application of Enhanced Oil Recovery (EOR) in oil and gas industry is very important to increase oil recovery and prolong the lifetime of a reservoir but it has been very costly and losing properties of EOR agent due to harsh condition. Nanoparticles have been used in EOR application since they are not degradable in reservoir condition and used in smaller amount compared to polymer usage. Commonly, EOR techniques are focusing on increasing the sweep efficiency by controlling the mobility ratio between reservoir fluid and injected fluid. Thus, this research aimed to analyze the nanofluid viscosity at different particle size and shape, volumetric concentration and types of dispersing fluid, as well as to determine the oil recovery performance at different nanofluid concentration. The nanofluid viscosity was investigated at nanoparticle sizes of 15nm and 60nm and shapes of 15nm spherical-solid and porous. Five nanofluid samples with concentration ranging from 0.1wt.% to 7wt.% were used to investigate the effect of volumetric concentration. Distilled water, ethanol, ethylene glycol (EG) and brine were used for the effect of dispersing fluids. Oil recovery was investigated at five different concentrations of nanofluid samples through flooding test. It was found that viscosity of nanofluid increased with decreasing particle size and increasing volumetric concentration. Solid shape particle and increasing dispersing fluid viscosity resulted in higher nanofluid viscosity. The higher the nanofluid concentration, the higher the oil recovery obtained. It can be concluded that nanofluid properties have been significantly affected by the environment and the particle used for potential EOR application.

1 Introduction

Enhanced Oil Recovery (EOR) has become the best solution to solve the problem of declining production and to prolong the life of a reservoir. By using this technique, the recovery performance for a reservoir can be increased up to 60% of oil compared to the conventional method which can only recover between 20-40% of oil [1]. EOR comes with several methods such as injection of miscible solvents and liquid carbon dioxide (CO_2) superfluid, polymer flooding, surfactant flooding, microbial injection and thermal methods. Besides that, another interesting method used is injection of nanofluids [2].

Nanofluid is a mixture of nano-sized solid particles; usually less than 100 μm and its base liquid as shown in Fig. 1. Examples of nanoparticles are metal oxides such as SiO_2, CuO, Al_2O_3, MgO and etc. while for the base fluids are water, brine, ethylene glycol, and etc. Nanofluid can be prepared in several ways such as single-step method and two-step method. One-step method is a process where preparation of nanoparticles and synthesis of nanofluids are done in a two-combined process while two-step method involves synthesizing of nanoparticles first and then mixing with base fluids [3].

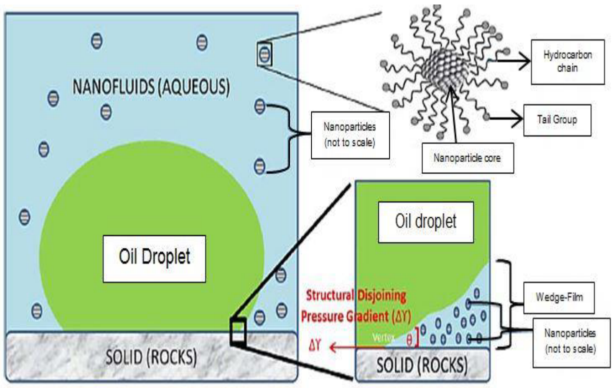

Figure 1. Structure of nanoparticle [3].

For oil and gas industry, one of the mechanisms of nanofluids that can be used for EOR method is viscosity alteration which is very important especially in recovering heavy oil. In addition, it can also be used to control the viscosity of displacing fluid (nanofluid) which then improves the mobility ratio thus increase the oil recovery. Sweep efficiency is influenced by mobility ratio (M) which is also dependent on the mobility of displacing fluid and displaced fluid (oil). The key of controlling the mobility ratio is by altering the viscosity of each one or both of the fluids [4]. This can be expressed in Equation 1.

$$M = \frac{\lambda_A}{\lambda_B} \quad (1)$$

where; λ = mobility = k/μ, k = permeability, μ = viscosity, A = displacing fluid and B = displaced fluid. The favorable value for mobility ratio is below than 1 while larger than that indicate low sweep efficiency [5]. The effect of mobility ratio is illustrated in Fig. 2. Basically, low mobility ratio is favorable to displace a fluid whereas high mobility ratio will reduce the sweep efficiency due to the "fingering effect" problem. In the industry, polymers are known to have the ability to increase the viscosity of a fluid but very costly compared to nanoparticles which required only in small amount due to its large surface area [6].

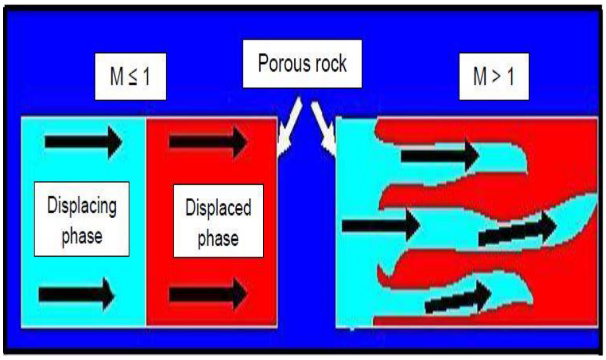

Figure 2. Mobility ratio effect on sweep efficiency [7].

Nanoparticles have been developed for various applications in EOR and can be summarized in three approaches, which are nanocatalysts, nanoemulsions and nanofluids [8]. The presence of nano-sized metal particles catalyzes the breaking of carbon-sulfur bonds within asphaltenes [9]. High viscosity provided by nanoparticle-stabilized emulsions can improve mobility ratio during the flooding, thus increase oil recovery. Mohd et al. (2015) have investigated the mobility of nanoparticle-stabilized CO_2 foam, who found reduction of mobility resulting in enhancing oil recovery [10]. Foam stability also increased with increasing nanoparticle concentration [11]. Zirconium oxide based nanofluids have been applied for wettability alteration, which have great potentials in changing oil-wet limestone towards strongly water-wet condition [12].

Several studies have been done to investigate the particle size effect on the viscosity of a nanofluid. Nguyen et al. (2007) found that the viscosity of nanofluid increased with increasing particle size in their studies on Al_2O_3-water nanofluid [13]. However, several contradictory results have also been obtained. A study by Lu and Fan (2008) on Al_2O_3-water and ethylene glycol (EG) for different nanoparticle sizes showed the decrease in viscosity with increasing particle size [14]. Another study using SiO_2 nanoparticles with sizes of 20, 50, 100nm also found that viscosity reduced with the increase of particle size [15]. There was limited research investigating the effect of particle shape on nanofluid viscosity. However, some studies found that the viscosity of nanofluid was greatly dependent on the particle shape. Timofeeva et al. (2011) reported that elongate particles like platelets and cylinders resulted in higher viscosity compared to spherical particles at the same volume fraction. Other shapes of nanoparticles are spherical, rod, sheet, etc. [16]. Based on previous researches, there was no contradiction about the effect of volumetric concentration on nanofluid viscosity. Das et al. and Putra et al. in 2003 proved that viscosity of nanofluid increased with increasing particle concentration [17, 18]. Thus, this paper aims to analyze the nanofluid viscosity with respect to nanoparticle shape and size, volumetric concentration and types of dispersing fluids, as well as investigating oil recovery at different volumetric concentration.

2 Methodology

Two major experiments have been conducted at room temperature and atmospheric pressure in order to meet the objectives of this research. The first part was the investigation of nanofluid properties and followed by oil recovery flooding test.

2.1 Preparation of nanofluid samples

The main materials used in this research were silicon dioxide (SiO_2) nanoparticles (spherical-solid shape of 15 and 60 nm as well as porous shape nanoparticle) purchased from US Research Nanomaterials Incorporation. These materials were used as they have been received. Paraffin oil, which was used to represent the real oil inside the sand pack for flooding test and Alpha-olefin Sulfonate (AOS) surfactant were provided by Chem Laboratory. Basically, the preparation of nanofluid for this research used two-step method, whereby nanofluids have been formed by dispersing solid nanoparticles into base fluid. Although aggregation of particles was the main challenge for this method, the problem has been minimized by adding some surfactant and application of ultrasonic. Viscosity tests were carried out using Brookfield DV-II+ Viscometer due to its suitability to measure the nanofluid viscosity. Ten readings were taken for every 30 seconds and the average of viscosity was calculated.

2.1.1 Effect of particle size

The nanofluid samples were prepared using SiO_2 nanoparticle. Two different sizes of SiO_2 nanoparticle used were 15 nm and 60 nm; dispersed in distilled water at concentration of 5 wt.%. To increase the nanofluids stability, 0.1 wt.% of surfactant was added and then sonicated for 15 minutes.

2.1.2 Effect of particle shape

The nanofluid samples were prepared using two different shapes of SiO_2 nanoparticle; spherical-solid shape and porous shape. Both samples were dispersed in distilled water at 5 wt.% concentration. 0.1 wt.% of surfactant was added and then sonicated for 15 minutes to increase the samples stability.

2.1.3 Effect of volumetric concentration

SiO_2 nanoparticle was dispersed in distilled water. Five samples of nanofluids were prepared with concentration of 0.1 wt.%, 0.5 wt.%, 2 wt.%, 5 wt.% and 7 wt.%. To retain fluid stability, 0.1 wt.% surfactant was added into each sample and then sonicated for 15 minutes.

2.1.4 Effect of types of dispersing fluid

Four samples of nanofluids were prepared using SiO_2 nanoparticles at concentration of 5 wt.%. Distilled water (DI), ethanol, ethylene glycol (EG) and also brine solution (30000ppm) were used as dispersing fluids with 0.1 wt.% surfactant and then sonicated for 15 minutes to prevent aggregation of particles.

2.2 Oil recovery flooding test

The flooding test was carried out to study the mechanism of nanofluid to recover oil in porous media. Sand pack was used as the porous media while paraffin oil to substitute crude oil. Paraffin oil was coloured with red dye to differentiate it with brine. The volume of oil recovered was divided to the total volume of oil inside the sand pack to measure the percentage of oil recovery. The oil recovery flooding test setup is shown in Fig. 3.

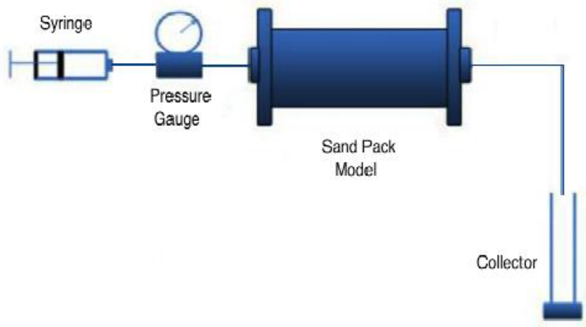

Figure 3. Flooding Test setup.

Sand pack was prepared using sieve shaker to get the uniform size of sand grain between 150-300 microns. The meshed sand was filled into the sand pack holder fully and tightly. The flooding setup includes the main components of sand pack, syringe pump, pressure gauge and measuring cylinder (collector). Firstly, the sand pack was fully injected with brine. Then, paraffin oil was injected into the sand pack until no water can be displaced anymore or called as irreducible water saturation. After that, the secondary recovery was applied to the system using brine which was injected at constant rate of 4 ml/min to recover the oil inside the sand pack. The oil recovered was collected in measuring cylinder and then calculated. Nanofluid was then injected into the sand pack to determine the recovery of EOR. For this purpose, different concentrations of nanofluids were injected at 0.1 wt.%, 0.5 wt.%, 3 wt.%, 5 wt.% and 7 wt.%. The injection was kept at constant rate of 4ml/min and the collected oil recovery in the measuring cylinder was calculated.

3 Results and discussion

3.1 Investigation of nanofluid viscosity

3.1.1 Effect of particle size

Fig. 4 shows the result of nanofluid viscosity versus particle size where the viscosity decreased as the particle size increased.

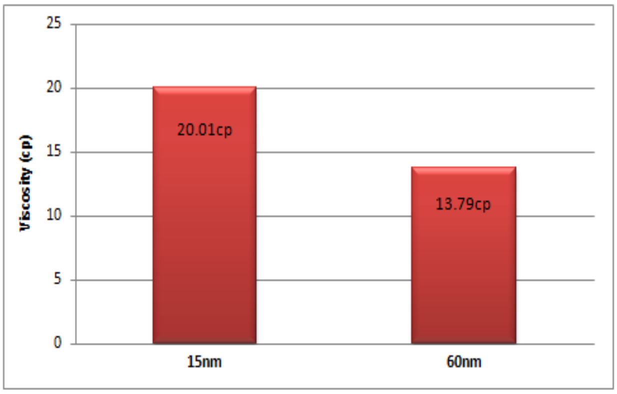

Figure 4. Viscosity versus particle size.

This was supported by finding from Chevalier et al. (2007), who measured the viscosity of SiO_2-ethanol for three different sizes of particle diameter 35, 94 and 190 nm [19]. Similar result was also found by Lu and Fan (2008) and Pastoriza-Gallego et al. (2011), who investigated the viscosity of CuO-water with sizes ranging 23–37nm [14]. According to Bryn McDonagh (2011), in a constant volume fraction, the number of particles increased when particle size decreased resulting in increasing interactions between particles, which then leading to an overall increase resistance to flow [20].

3.1.2 Effect of particle shape

Fig. 5 illustrates nanofluid viscosity versus particle shape, whereby spherical-solid shape provided higher nanofluid viscosity than the sample with porous shape.

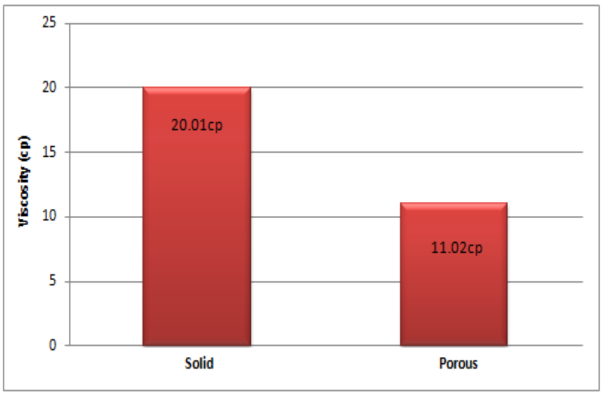

Figure 5. Viscosity versus particle shape.

According to Timofeeva et al. (2009), elongate particle such as platlets and cylinders resulted in high nanofluid viscosity compared to spherical shape [16]. As porosity reduced the resistance of fluid to flow during in contact with surface, hence the viscosity was reduced too [21, 22]. Based on this statement, it can be concluded that the result was acceptable as the viscosity for sample with porous shape was lower than the sample with spherical-solid shape. Although there were not many researches focusing on particle shape, this parameter can be a strong factor in manipulating the viscosity of nanofluid to suit the requirement of its application.

3.1.3 Effect of volumetric concentration

From Fig. 6, it can be clearly seen that the viscosity of nanofluid increased as the volumetric concentration increased. In lower concentration, low amount of particles were inside the water, thus less resistance for the fluid to flow resulting the flow property of water to be dominating. In contrast, for higher concentration solution, there were higher amount of particles, which resulted in more resistance as the particles collided each other and reducing the original flow property of the water; higher viscosity [23]. This result was supported by several researchers, who found identical results. For example, Das et al. (2008) reported that increasing Al_2O_3-water nanofluid concentration has resulted in increasing nanofluid viscosity [24]. Nguyen et al. (2007) concluded that particle size effects are more significant for high particle volume percentage. Their studies on Al_2O_3-water nanofluid exhibited that for volume concentrations below 4%, viscosities corresponding to 36 nm and 47 nm particle size were almost equal. In contrast, for higher concentrations, viscosities were very different [13].

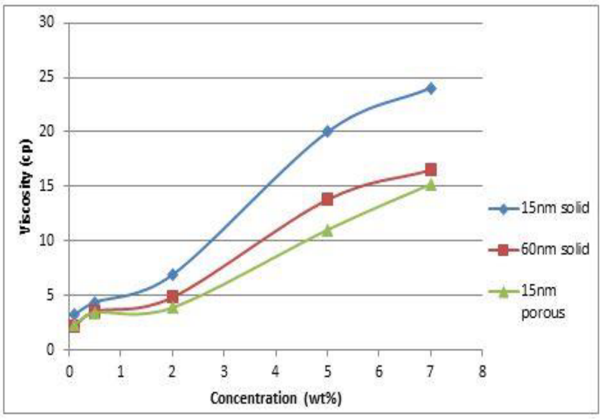

Figure 6. Viscosity versus volumetric concentration.

3.1.4 Effect of dispersing fluid type

Fig. 7 shows the average viscosity of nanofluid samples dispersed in four different fluids. The average viscosities were found to be 20cp, 36.3cp, 19.52cp and 20.43cp for the samples using DI, EG, ethanol and brine, respectively. The highest viscosity was achieved by using EG as the dispersing fluid while the lowest was when using water. These results were affected by the viscosity of the dispersing fluid itself as the viscosity of EG and ethanol were approximately 20cp and 1cp, respectively at room temperature. Thus, it can be concluded that the viscosity of nanofluid was dependent on the viscosity of its dispersing fluid.

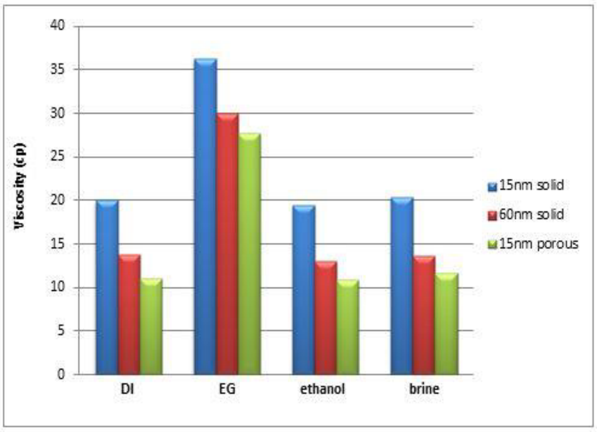

Figure 7. Viscosity versus type of dispersing fluid.

The average viscosity for samples using DI and brine were almost the same to the sample using water as the original viscosities for the three dispersing fluid were nearly the same. The investigation of nanofluid viscosity with respect to different dispersing fluid is important as the property of dispersing fluid could be one of the factors to influence the nanofluid produced. Besides that, further studies of dispersing fluid could be beneficial to enhance the application of nanofluid. Selection of dispersion fluid used was based on several researches such as in a study of Al_2O_3-water and ethylene glycol (EG) for different volume fractions and in a study of SiO2-ethanol with three different nanoparticle sizes [19, 14].

3.2 Investigation of oil recovery

Fig. 8 shows the result of oil recovery based on varying nanofluid concentration. From the figure, the oil recovery increased as the concentration of nanofluid increased.

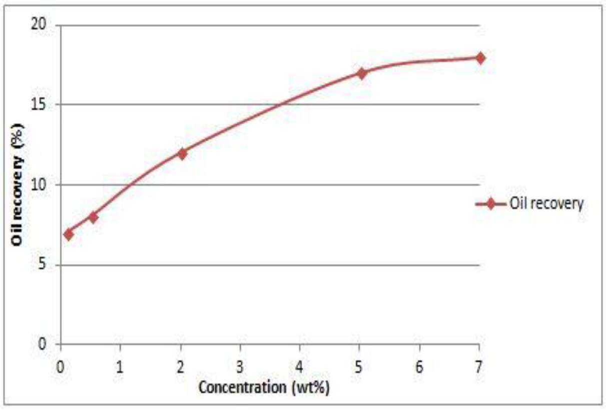

Figure 8. Oil recovery versus nanofluid concentration.

The lowest oil recovery was obtained from the injection of 0.1 wt.% of nanofluid with percentage of 7% recovery while the highest recovery was from the injection of 7 wt.% of nanofluid with 18% recovery. However, the injection of 7 wt.% of nanofluid might contribute to some problems due to higher precipitation of nanoparticle. According to Hendraningrat et al. (2013), high nanoparticle concentration and injection rate could lead to high impairment. The retention of nanoparticles inside the core plugs induced porosity and permeability with some blockage to reservoir hence reduced the porosity [25].

From this experiment, small difference of oil recovery (1%) was found between 7 wt.% and 5 wt.% of nanofluid. Furthermore, the precipitation rate for 5 wt.% sample was low, indicating small effect on the reservoir porosity. By this consideration, it can be concluded that 5wt% nanofluid was the most effective concentration to be used in recovering oil as it could provide high oil recovery, low precipitation rate and less impact to the reservoir properties. For this flooding test, only SiO_2 nanoparticle with size of 15nm and spherical-solid in shape was chosen in preparing the nanofluid samples. The selection was based on previous viscosity tests, which resulted in highest viscosity and DI as the dispersing fluid by considering the economic benefit for industrial application.

This result also proved the Equation 1 which theoretically explained that high viscosity of displacing fluid reduced the mobility of that fluid, resulting in mobility ratio less than one [4]. Fig. 8 shows the highest percentage of oil recovered was from the highest concentration of nanofluid sample, and vice versa. This was due to 7 wt.% nanofluid concentration had the highest viscosity which could recover the paraffin oil inside the sand pack more effectively. Maintaining low mobility ratio is very important to increase the sweep efficiency for recovering more oil and minimizing 'fingering effect' [5, 7].

4 Conclusion

The present study has revealed that the application of EOR can help to increase the total oil recovery and prolong the life of reservoir. From this experiment, it can be concluded that viscosity of nanofluid increased as the particle size decreased, volumetric concentration increased, using of solid shape particle instead of porous shape and also using high viscosity of dispersing fluid. From the flooding test, the percentage of oil recovery increased with increasing concentration of injected fluid but too high concentration could damage the reservoir productivity. The outcomes could be very useful for industrial application as it provides new knowledge on nanofluid properties with respect to particle shape and size, volumetric concentration and types of dispersing fluids to fully optimize the potential of nanofluid properties. This could be potentially applied in nanofluid flooding of EOR application.

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