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Influence of Nano Fluid on Interfacial Tension oil/water and Wettability Alteration of limestone

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Abstract. Nano materials have a great potential for enhanced oil recovery applications. The present research investigated the effect of nanoparticles on interfacial tension and wettability alteration. The silica oxide nanoparticles with different size 52, 65 and 5nm were used. The wettability alteration was evaluated by dipping the limestone rock in different concentrations of Nano silica (0.01-1wt. %) for a certain silica size suspended in salinity water. Measurements performed in three phase system (oil/water/solid) using pandel droplet and sessile drop method respectively by opitical- tensiometer. Results showed that Nano fluid can significantly reduce contact angle and alterated rock wettability with increasing Nano-silica concentration due to their high surface area in which make it more reactive, yielding the best results when using 0.01 wt.% Nano silica that decreased contact angle to (29°, 59°, 69°, 26°) for 10, 52, 65nm Nano silica size and modified Nano silica respectively for gas /liquid/ solid system. in addition, for liquid /liquid/solid system, 0.01 wt.% Nano silica was decreased contact angle to (32°, 36°, 41°, 30°), interfacial tension to (2.26, 2.8, 3, 3.6Nm) for 10, 52, 65nm Nano silica size and modified Nano silica respectively, Interfacial tension decreased with increasing Nano silica concentration, but NPs size have slightly effect on interfacial tension.

Keywords: Carbonate; EOR; Nano Silica; Wettability alteration

1. Introduction:

The residual oil is trapped in reservoir pores due to interfacial and surface forces. This trapped oil recovered by decreasing capillary forces that prevent oil from flowing within the pores of reservoir rock and into the well bore. Capillary pressure which is the force necessary to thrust oil droplet through a pore throat reduces by decreasing of oil/water IFT and wettability alteration [1].

During enhanced oil recovery (EOR) process, the presence nanoparticles in injection fluid reducing interfacial tension with or without surfactant where the interfacial tension is one of the important mechanisms for mobilizing the residual oil[2]. Adsorption of nanoparticles onto the surface of the fluid will effectively reduce the interfacial tension between those fluids [3]. The magnitude of interfacial tension reduction is directly related to nanoparticle concentration, with higher concentration favorable for lower IFT [4].

Small size of NPs makes it probable to push the residual oil in the small pores that unrecovered in polymer injection (named Inaccessible Pore Volumes). Despite of continuous fluid bulks, there is another advantage about dispersed particles; dispersed particles can hit the porous media wall and remove the oil on the wall.

Abbas et.al.,2015 [5] investigated a special type of Nanoparticles named Polysilicon. Hydrophobic and lipophilic polysilicon and naturally wet polysilicon as EOR agents in water-wet sandstone rocks. The results showed a change to water-wet condition and intensive decrease in oil-water interfacial tension from 26.3 mN/m to 1.75 mN/m and 2.55 mN/m after application of HLP and NWP Nano-fluids respectively. As a result, oil recovery increased by 32.2% and 28.57% when a 4g/lit concentration of HLP and NWP Nano fluids are injected into the core samples respectively.

Li, et al.(2013) [6] Studied the effect of silica Nano fluids and salt-water injection and found that the amount of intake with Nano-fluid injection increased due to decreasing in interfacial tension but adsorption of nano-particles have to be considered.

Mohammadi, et al. (2014)[7] investigated the effect of gamma-aluminum NPs on wettability of limstone rocks and by the flooding tests for the third time they found that aluminum Nano fluids injection in a 0.5% wt increased oil recovery to 11 %.

Yousefv and and Jafari(2015)[8] Studied the presence of polymer and NPs on oil recovery. They found that EOR in the presence of materials increased to 10 %. Alomair, et al.(2015) examined various NPs to find proper Nano fluids for EOR and they found that largest reduction in interfacial tension due to high surface area of NPs, and by increasing the concentration of NPs, the interfacial tension is reduced to a greater extent.

2. Experiment:

2.1 Materials, Chemicals and instrument

Carbonate Rock from mishrif formation-Iraq and gas oil from aldora refinery was supplied , Sand was supplied from Al-Anbar-Iraq with 99% silica oxide. Furthermore, Sulfuric acid 98%, Barium chloride 99% anhydrous, and Nano silica (5-15nm) purity 99.99% were supplied from sigma Aldrich. Company. Also, Sodium chloride 99.5% from alpha company, and Sodium hydroxide were supplied

2.2 Preparation of Nano silica

Sand was crashed by ball miller and sieved to 47µm and 37µm. From each size, 40g of sand was mixed with 100g sodium hydroxide. The two samples heated to 500 °C for 30 min. where sodium hydroxide melts coated the sand particles, making the mixture a grayish/white color of Sodium Silicate as shown in equation (1), then samples cooled to room temperature.

$$2NaOH_{(l)} + SiO_{2\,(s)} \rightarrow Na_2SiO_{3\,(s)} + H_2O_{(g)}. \quad (1)$$

Distilled water was added with mixing until sodium silicate completely dissolved, where salicylic acid and sodium oxide formed as shown in equation (2).

$$2Na_2SiO_{3\,(s)} + 4H_2O_{(l)} \rightarrow 2H_4SiO_4 + 2Na_2O. \quad (2)$$

The solution was acidified (pH=1) to precipitate Nano silica by addition of concentrated sulfuric acid. Coagulated Nano silica was separated from the solution by vacuum filtration and then washed with distilled water to remove sulfate ions which controlled with 6% BaCl2 solution. The separated cake was dried in oven at 100C° for 48hr.

2.3 Nano fluid synthesis

Nano fluids were prepared in two-steps. First, a certain amount Nano silica was added to the Salinity water (9wt% NaCl, pH 6.5) under mixing condition to obtain concentration from 0.01 to 1wt. %. Second, suspended silica was subjected to sonication using an ultrasonic mixer for 15 min. with high energy to avoid agglomeration.

2.4 Rock sample preparation

Carbonate rock Slices were prepared by cutting and polishing, and then dried at 100 °C for 1hr. The carbonate slices as shown in figure (1) were dipped in a 20 ml of different Nano fluid concentrations (0.01-1wt. %) for 1hr under vacuum circumstance. Then slices were dried at 100°C for 1 hour.

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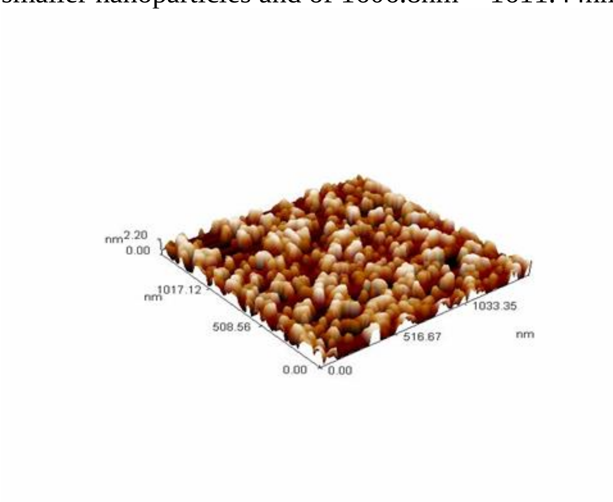
Figure 1. carbonate slices dipped in Nano fluid

2.5 Nano silica characterization

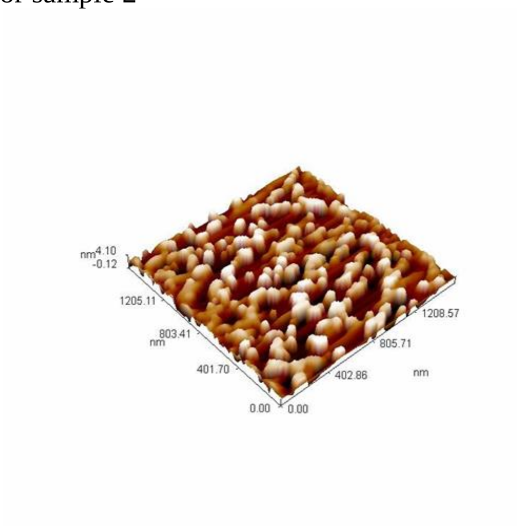
Atomic Force Microscope (AFM).

The Atomic Force Microscope (AFM) allows for 3D characterization of NPs with sub-nanometer resolution.

A 3D rendering type of the surface topography was presented in Figure 2a and 2b respectively with good resolution scan was achieved for a surface of 3051.48nm × 1550.41nm for sample 1, as it has smaller nanoparticles and of 1606.8nm × 1611.44nm for sample 2



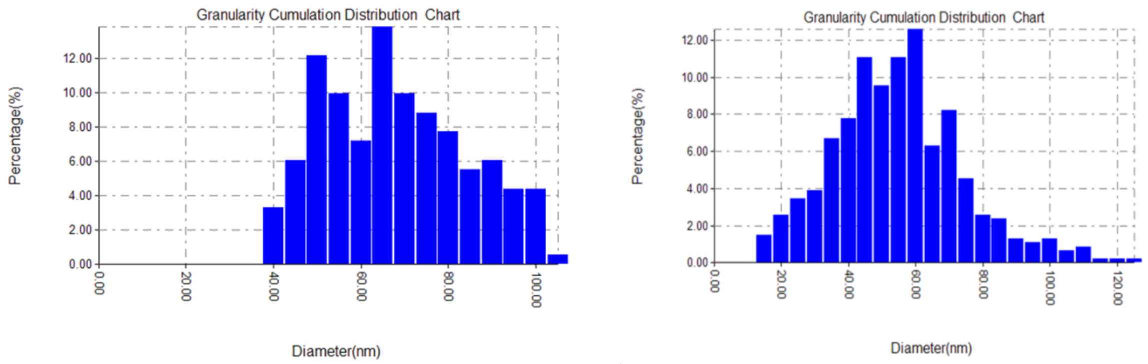
(a)



(b)

Figure 2. AFM 3D images in topography for (a) avg. particle size 65nm (b)) avg. particle size 52nm

Using data from image analysis software module, a histogram of the NPs distributions was evaluated as shown in figure 3. Figure 3a and 3b showed that the average particle size was 65nm and 52.4nm for 47 and 37 μm sieved sand. Also showed that the particle size distribution is not quite a perfect Gaussian line, where the smaller number of objects that were present on the surface and were therefore subject of the statistical analysis



(a) (b)

Figure 3. relative size distribution built up from AFM image for (a) sample 1 (b)

Accelerated Surface Area and Porosimetry System.

The Micromeritics ASAP 2020 Plus integrates a variety of automated gas sorption techniques into a single as shown in figure (4). The system provides high-quality surface area, porosity, chemisorption and physisorption isotherm data to materials analysis laboratories with ever-expanding analytical requirements. Physical adsorption of nitrogen molecules on a solid surface analysis was applied for the measurement of the specific surface area of prepared Nano silica. For silica particle size 52nm, the Surface Area: 474.9429 m^2/g also Nano silica with 65nm have surface area of 440m2/gm.

Wettability alteration test

Theta Lite optical tensiometer is accurate meter for simple measurements of contact angle and surface free energy as shown in fig.2. It also measures surface and interfacial tension.

Wettability was calculated by measuring contact angle between liquid drop and rock sample in oil medium for several systems. The contact angle measurements were repeated as mentioned before for each system. Furthermore interfacial tension was measured using pandet drop method between Nano fluid and oil in room temperature condition.

3. Result and Discussion

Porous rocks saturated with different liquid are a complex system of constant interactions between all existing fluids and between fluids and rock. Wettability is the tendency of liquid to spread on a solid surface in the presence of another immiscible-liquid. This is due to the interfacial tension between the current fluid phases and their individual adhesion to solids. The wettability of the pore wall depends on the chemical composition of the liquid and the composition of the rock metal.

3.1 Contact angle

Figure 6 showed that the effect of Nano-particle concentration on wettability alteration. In which contact angle decreased with increasing the concentration gradually that from 0.1-1wt. % Nano-particle, decreasing in contact angle becomes approximately smaller.

Nanoparticle size effect on contact angle is Attributable to the surface energy, in which surface area increased with smaller sized nanoparticles, which lead to increase the surface free energy so that contact angle decreased. Other factors like roughness of the surface and distribution of nanoparticle on the surface which can be also control contact angle. The difference in the thickness of adsorption layer of the nanoparticles on the surface for different NPs size may causes deviation in the contact angle values, particularly for intermediate sized nanoparticles [9].

Addition of Nano silica to the saline water caused abrupt reduction in contact angle from 72□ to under 50□ for different size of NPs., in which contact angle gradually decreased with increasing NPs concentration until reach to approximately constant value as shown in figure (4). Also, it is interesting to note that particle size inversely proportional to the surface area /volume ratio, which reduce in the ability to perform surface interactions [10]. Because the smaller size, technically more chemically reactive, it appears that a mechanical, rather than a chemical, mechanism is the driver for enhanced oil recovery.

Nanoparticles adsorbed on the surface of the rock promoted oil displacement thus contributing tothe inevitable change in wettability from oil-wet to water-wet in a very efficient way

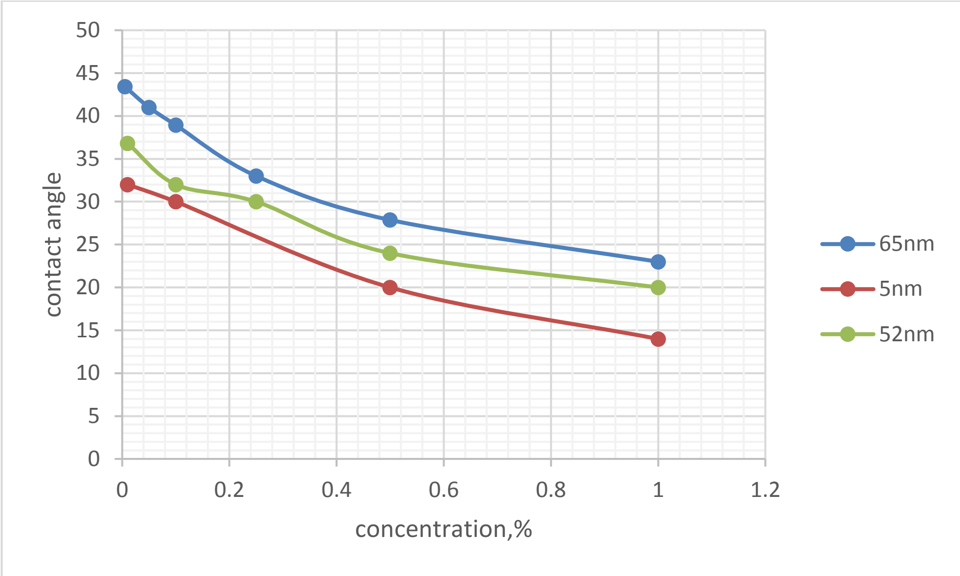


Figure 4. contact angle in degree versus Nano silica concentration for different size of NPs

3.2 Spreading

Wetting and spreading are the important key in oil recovery the distinction between the different wetting states is usually made by considering the spreading coefficient, which represents the surface free energy relative to its value for complete wetting.

$$S = \gamma_{SO} - (\gamma_{SW} + \gamma) = \gamma(\cos\theta - 1) \quad (3)$$

Note that for complete wetting the equilibrium spreading coefficient is zero. The solid-vapor interface then consists of a macroscopically thick wetting layer, so that its tension is equal to the sum of the solid-liquid and liquid-vapor surface tensions[11].

Figure (5) Silica NPs concentration directly related with spreading coefficient, in other wise size of NPs inversely related to spreading due to intermolecular force (surface tension) which contract the surface, in which it is responsible for the shape of liquid droplets. In practice, external forces such as gravity deform the droplet [12].

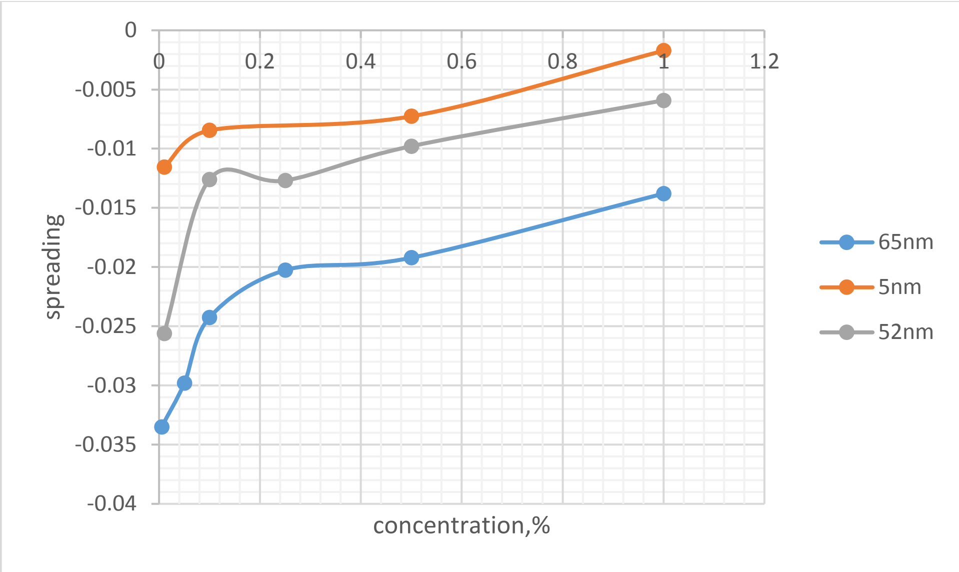


Figure 5. spreading versus concentration for different NPs

3.3 Interfacial tension

Oil-water interfacial tension was measured before and after application of Nano fluids in ambient conditions, that have a great potential for increasing pore scale displacement efficiency through reduction of interfacial tension and wettability alteration. Figure (6) showed a great reduction in interfacial tension. In other hand, it had been show that different NPs size approximately the same effect on interfacial tension

Decreasing interfacial tension will make it easier for oil droplets to move through pore throats by decreasing work of deformation needed. Reducing interfacial tension rising the capillary number. A higher capillary number causes an increase in oil displacement efficiency

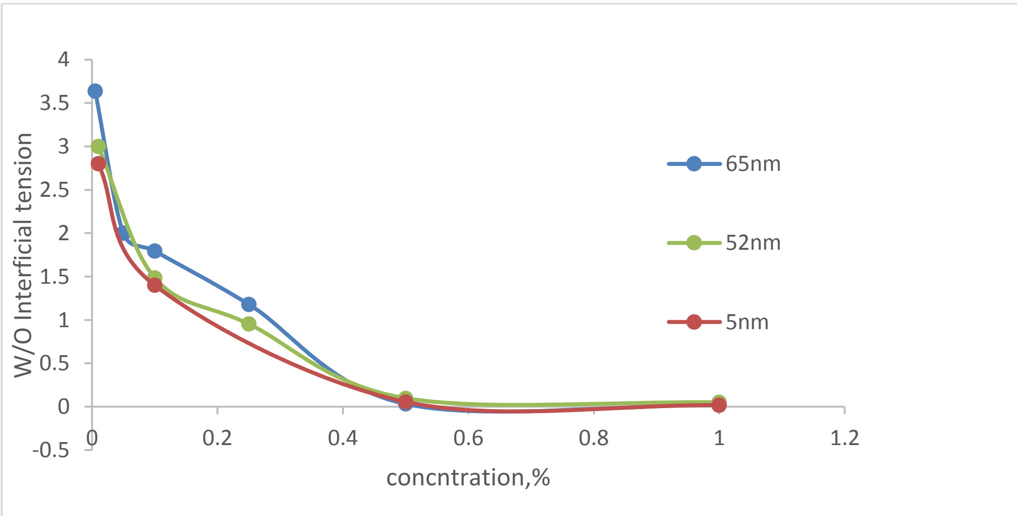


Figure 6. W/O interfacial tension versus Nano silica for different size of NPS

4. Conclusion

All Nano fluids used in this research improve oil recovery through two major mechanisms of interfacial tension reduction and wettability alteration to less oil-wet condition. However, these Nanoparticles have different level of contribution in Reduction of interfacial tension and alteration of rock wettability. Nanoparticles with size 5nm have stronger impact on rock wettability while all sizes

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Nanoparticles have approximately the same influence on reduction of oil-water interfacial tension. Also the results showed that the minimum value NPs concentration for optimum interfacial tension and contact angle was 0.01%.

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