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INVESTIGATION ON NANOPARTICLES' EFFECT ON INTERFACIAL FORCES FOR ENHANCED OIL RECOVERY

Yusuff Afeez[1], Noorhana Yahya[1], Chai Mui Nyuk[1], Bilal Al-qaseem[1], Saima Qureshi[1], Sikiru Surajudeen[1], Ali Abdullah[2], and Wahab Fathai[1]

[1] *Department of Fundamental and Applied Sciences, UTP, Malaysia*
[2] *Department of Geology, Bayero University, Nigeria*

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Abstract

The preferred objective of oil industry is to lower the interfacial forces impeding fluid flow and efficiency of injected fluid. This has paved the way for exploration of nanoparticles as suitable candidate for this goal. Although many research activities have been done with nanoparticles, the interest of looking for nanoparticles that can reduce interfacial tension and contact angle at high reservoir temperature condition is rare. Therefore, in this paper, molecular dynamics simulation study was first employed to determine indirectly the influence of different dielectric nanoclusters on contact angle using stress tensor at 120°C. It was found that ZnO nanoparticles show highest reduction in stress up to 60% of the referenced oil-brine value. Subsequently, experimental investigation of zinc oxide nanoparticles on Interfacial tension at both 25°C and 95°C were conducted. By varying concentration, 0.05 wt% was observed as optimum concentration and further used for high temperature application. The results conclude that zinc oxide is a temperature-oriented nanoparticle as its capable of reducing both the stress acting along oil-rock interface and oil-brine interfacial tension at high temperature.

***Keywords**: Interfacial tension; Contact angle; Nanoparticles; Enhanced oil recovery; Molecular dynamics simulation.*

1. Introduction

Different enhanced oil recovery methods have been continually emerging as a result of inefficiency of water or pressure drive recovery method. They are formulated to alter the surface properties of reservoir components that may facilitate production of residual oil trapped in the reservoir [1-2]. The main cause of failure for displacing fluid is the capillary pressure(stress) arising from the interfacial forces between solid/liquid or liquid/liquid interface [1]. Therefore, the goal of any enhanced recovery techniques is either to increase the viscosity of the injected fluid or lower the interfacial forces as expressed in equation 1 below:

$$N_c = \frac{Viscous\ forces}{Interfacial\ forces} = \frac{\mu v}{\sigma_{ow} cos\theta} \qquad (1)$$

where N_c is the capillary number; μ= the dynamic viscosity of the displacing fluid; v= velocity; θ= oil/ water contact angle(wettability); σ_{ow}= interfacial tension (IFT) between oil and water (dyne/cm).

The capillary number is a dimensionless parameter which is directly proportional to the productivity of oil by any developed method. As the preferred option for oil industry is to reduce the interfacial forces due to the high cost associated with viscosity increment of the injected fluid, therefore the focus of this paper is on the reduction of interfacial forces. Surfactant has been historically used to achieve this due to their ability to adsorb at the interface of two immiscible fluids and consequently lower the interfacial tension between oil and water [3-4]. However, due to their instability at high temperature reservoir condition, they degrade and eventually unable to achieve their desired feature [5-7]. This is similarly the limitation of chemical based EOR method [8]. Thus, there is a need to look for alternatives that will be suitable for high temperature conditions.

The attractive features of nanoparticles such as thermal stability, low cost of fabrication, high surface to volume ratio, high adsorption affinity and their ability to penetrate deep into the reservoir make the use of nanotechnology a vibrant, competitive and economical alternative to conventional recovery methods. Hence, the use of nanomaterials for oil recovery has gained increased attention with many different mechanisms proposed to be responsible for increase in their productivity. Despite the fact that many laboratory reports have been made on nanoparticles proven ability to enhance the sweeping efficiency of water flooding by reducing oil-brine IFT or wettability alteration [7,9-11], the choice of looking for nanoparticles that can influence both interfacial forces parameters at high temperature condition is not common. Therefore, in this paper, ten different dielectric nanoparticles were investigated for this purpose. Due to high capital involvement and technical limitation of equipment in their scope of operation, two methods were employed; molecular simulation study and experimental investigation for contact angle and interfacial tension reduction respectively.

2. Molecular simulation study

2.1. Theory: stress autocorrelation function (SACF)

In molecular dynamics (MD) simulation, stress autocorrelation function is a summative function that can be used for estimating the stress between two surfaces. Therefore, it takes into account the total effects of all atoms involved in the process [12] and can be expressed as follows:

$$C_{xy}(\mathrm{t}) = \langle \sum_{x<y} P_{xy}\,(t)\, P_{xy}\ \ (0)\, \rangle \qquad (2)$$

where P_{xy} refers to an independent component of the pressure (stress) in the xy direction. For a molecular fluid, two formalisms are employable for the estimate of stress tensor with either method suitable: atomic formalism and molecular formalism with no much variance in result obtained [13-15]. The difference being that in atomic method, the stress tensor is calculated based on the motion of individual atoms constituting each of the molecules in the system as illustrated below:

$$P^{(a)}\,\mathrm{V} = \sum_{i,a} m_{ia}\, v_{ia}\, v_{ia} + \ \sum_{i,a} r_{ia}\, f_{ia} \qquad (3)$$

where m_{ia}, v_{ia} r_{ia} and f_{ia} are the mass, position, velocity, and force on atom of a molecule I.

For the molecular formalism, the stress tensor calculation is based on the motion of the molecules in the system, given as:

$$P^{(m)}\,\mathrm{V} = \sum_i m_i\ v_i\ v_i + \sum_i r_i\, f_i \qquad (4)$$

where $m_{i,}\, v_i$, r_i *and* f are the mass, centre of mass position, centre of mass velocity, and the total force on molecules i.

Thus, SACF is employed to estimate the stress between oil and rock surface with and without nanoparticles. The lower the stress, the higher the oil mobility and consequently reduces contact angle.

2.2. Methodology

In this study, the model of reservoir was built using material studio produced by Accelrys. Periodic boundary condition was imposed on the generated rock-particles-fluid structure while adopting COMPASS force field as the simulation force field. This is so because of its suitability for analysing both organic and inorganic molecules, metal oxides, polymers etc in condensed matter application [16]. The COMPASS force field can therefore be accurately used to determine the interaction parameters in heterogeneous fluid-particles system. Ewald and atom based summation methods were selected for computing non – bonded interactions; coulomb electrostatic and van der wall force respectively. The model of the 10 nanoclusters investigated ((zinc oxide (ZnO), silica oxide (SiO_2), titanium oxide (TiO_2), aluminium oxide (Al_2O_3), tin oxide (SnO_2), barium titanate (BaTiO3), zirconium oxide (ZrO), nickel oxide (NiO), magnetite (Fe_3O_4), molybdenum disulphide (MoS_2)) were constructed first, spherical in shape and 3$\dot{A}$ in radius. Thereafter, each was geometrically optimised for lowest energy configuration. The sandstone reservoir component was represented by quartz-alpha, as it is the major constituent of this rock. The model was placed inside a supercell to make the surface closer to reality. Furthermore, vacuum slab construction was made with a thickness of 50Å to limit non-bonded interactions between fluid/particles and rock surface. After this, the oil component, n-hexane construction ensued, followed by brine (water and NaCl) and the final reservoir model was built using Amorphous module. The simulated oil reservoir model was constructed without nanoparticles first and later on with each type of nanoparticles as presented in Fig. 1. All simulated structures were equilibrated before production runs. For annealing, 50,000 thermal cycles were chosen between 300 and 500 K. The NVT (constant number, volume and temperature) and NPT (constant number, pressure and temperature) were integrated using 200 ps and 300 ps – respectively [16-17] with Anderson-thermostat and Berendsen-barostart [18].The NVT ensemble was used to bring the system to the desired temperature (120°C) while production run was done in the NPT ensemble.

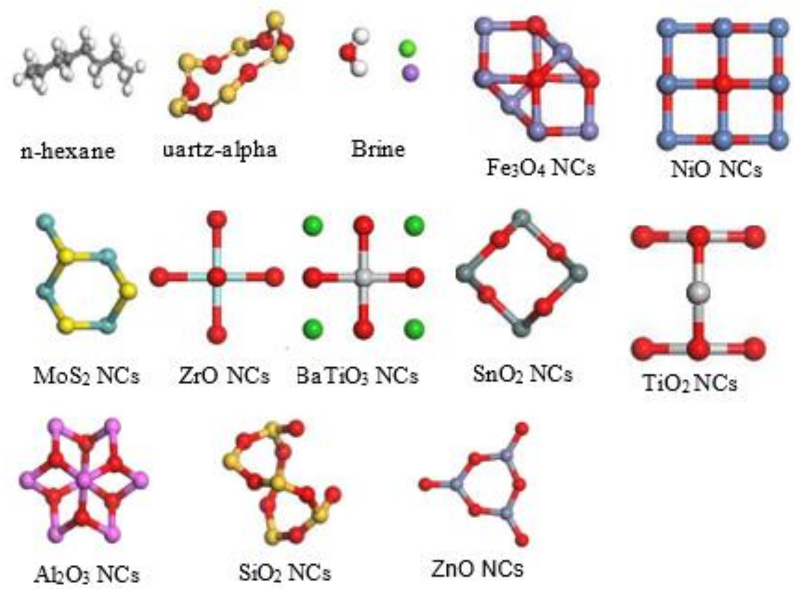


Fig. 1. Images of constructed models (a) individual reservoir components and investigated nanoclusters (NCs)

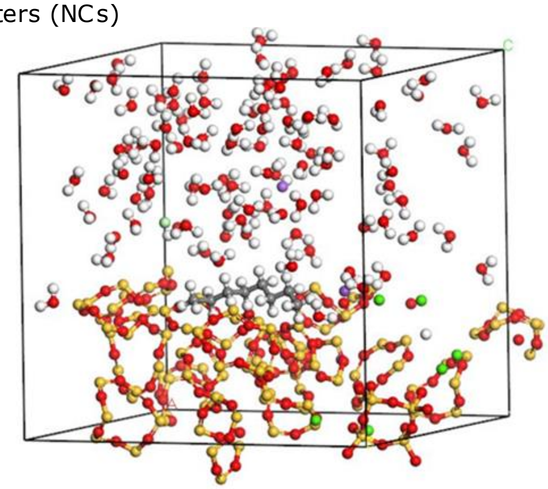


Fig. 1. Images of constructed models (b) Reservoir model without NCs

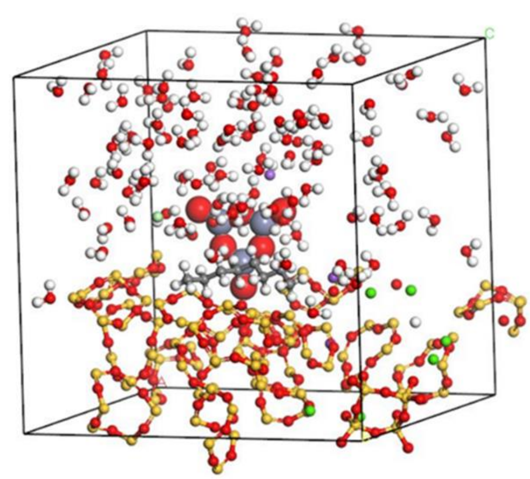


Fig. 1. Images of constructed models (c) Reservoir model with NCs

3. Experimental study

As a result of zinc oxide capability to reduce the surface stress the most at high temperature based on our simulation findings, it became desirous as well to study the effect of this nanoparticles on interfacial tension of oil and water at ambient and high temperature condition. This is imperative because interfacial tension is the second component of interfacial forces accountable for increasing capillary number, as given by Equation 1. Therefore, zinc oxide nanoparticles were synthesized for further investigation.

3.1. Synthesis of zinc oxide NPs

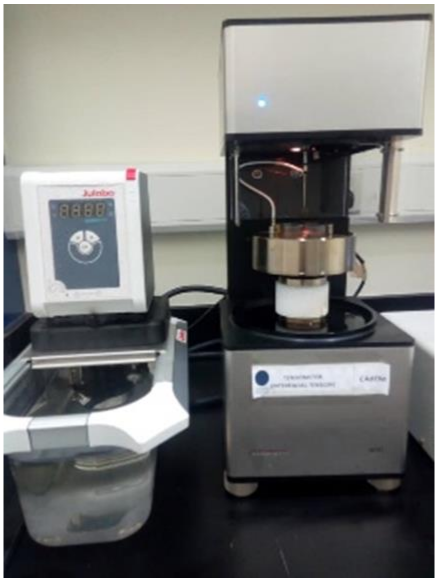
Zinc oxide nanoparticles had been synthesized through wet- chemical sol-gel route using zinc nitrate, citric acid, and distilled water as done by Soleimani *et.al.* [7] with modifications. All chemicals were obtained from Sigma Aldrich, at high purity grade. Zinc nitrate and citric acid were formulated with a mole ratio 2:1 respectively. Zinc nitrate represent the precursor, distilled water as hydrolysing agent and citric acid as a catalyst. Firstly, the solution of zinc nitrate (74.37 g) and citric acid (52.54 g) were prepared with distilled water (100mL) separately. Thereafter, citric acid solution was mixed with zinc nitrate solution and manually stirred for about 30 minutes before transferring to hotplate where it was stirred magnetically and heated up to 90°C until gel formation. The produced gelatin was dried in an oven for 48hrs and nanopowder was obtained. After pulverizing the nanopowder mechanically with mortar and pestle, the particles were calcined at 320°C for phase improvement and total desiccation. In order to affirm the integrity of the as-synthesized nanoparticles, FESEM with EDX characterizations were done (Figure 4). The elemental constituents of the analysed sample were obtained from EDX analysis using Zeiss Supra 55 VP.

3.2. Preparation of nanofluid

Two steps-method was adopted for the preparation of zinc oxide nanofluid (NF). The NPs were prepared using brine as base fluid with 11000 ppm NaCl concentration and stirred magnetically for 1½ hrs for perfect distribution. As a result of agglomeration concern which is associated with nanoparticles over time, the NFs were further sonicated with ultra sonicator probe at ambient condition for 1 hrs to obtain stable and homogeneous suspension without additional use of any stabilizer.

3.3. Interfacial tension measurements

The dataphysics Tensiometer was used to determine the interfacial tension between Oil and brine first at the two operating thermal conditions (25°C and 95°C) first followed by nanofluid at varying concentration. The 95°C was chosen in contrast to 120°C simulated temperature due to the limitation of the water bath temperature control equipment placed beside tensiometer as illustrated in Figure 2.



Initially, the measuring vessel, of 70 mm in height, was filled with light phase (crude oil) to more than one-third, and placed inside Tensiometer cell for first reading without touching the test body, the Wilhelm plate. (Note: The Wilhelm plate was repeatedly cleansed and sterilized for each of the two phases involved in the measurement). After a while, the vessel was exchanged with that containing dense phase and fraction out of the previous vessel's fluid was added and final IFT readings were obtained. Each reading was repeated three times and then averaged for the whole concentration tested.

Fig.2. IFT measuring set up (a) water bath temperature control (b) dataphysics Tensiometer

4. Result and discussion

4.1. Simulation study

The examination of nanoparticles effect on the stress between solid/liquid interface with MD simulation at high temperature condition is presented. Figure 3 shows the simulation result of stress autocorrelation function (SACF) between oil-rock with and without nanoparticles (NPs). As the mobility of a fluid over a surface increases when stress reduces, therefore, the lower the stress between oil and rock, the lower its contact angle and consequently enhanced displacement. The first 8 NPs were able to reduce the surface stress. While nickel oxide NCs shows no effect, molybdenum disulphide NCs caused increase in stress.

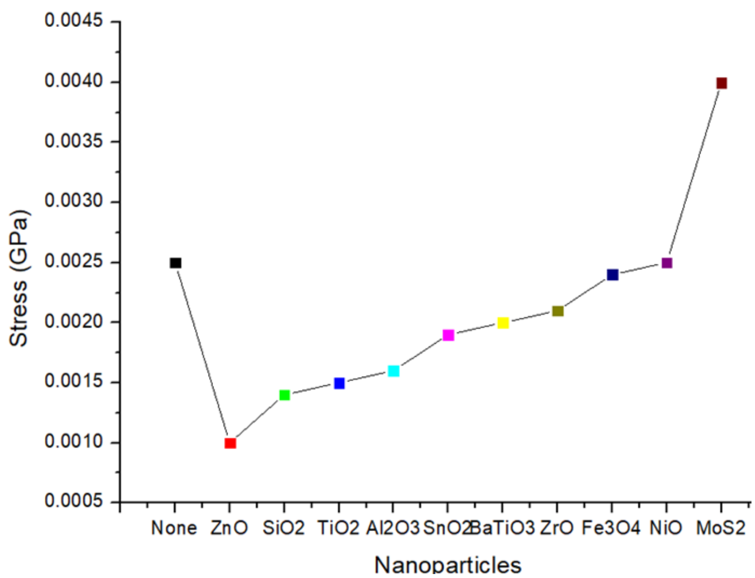


Fig. 3. Graphical result of surface stress obtained between oil/rock interface with and without NPs

This observation can be explained in terms of electrostatic interaction and adsorption energy of the NCs. When a particle bears opposite charge to the interacting surface, they attract each other and vice-versa. Depending on the degree of inherent potential, surface charge density and adsorption energy of adsorbate on the sorbate, the surface properties of the substrate could be altered. As the surface of sandstone rock is negatively charged (SO-) inside electrolytic solution, it can undergo electrostatic interaction with charged surface. Most nanoparticles also developed a surface charge at their interface when immersed in electrolyte solution. Nickel oxide has zero charges on its surface while molybdenum disulphide is negatively charged also, this caused its negative effect while nickel oxide lack of surface charge can render it unreactive to be able to bring any appreciable result. Among the nanoparticles that are able to reduce the stress, zinc oxide has the highest reduction of about 60% of the referenced stress value. Its positively charged surface, temperature sensitiveness and high adsorption energy could trigger the alteration effect of ZnO NC

4.2. Experimental study

4.2.1 Field emission scanning electron microscope (FESEM + EDX)

The synthesized ZnO nanoparticles were characterised using Field emission scanning electron microscope (FESEM) to determine its size and shape, as shown in Figure 4. As observed, the NPs form distinct and well-formed clusters of spherical nanostructures with globular terminations. The NPs are uniformly sized with an average size of 35 nm. Figure 5 is the graphical result obtained from the Energy Dispersive X-ray analysis with elemental distributions. This confirm the nature and purity of the synthesized nanopowder to be zinc oxide

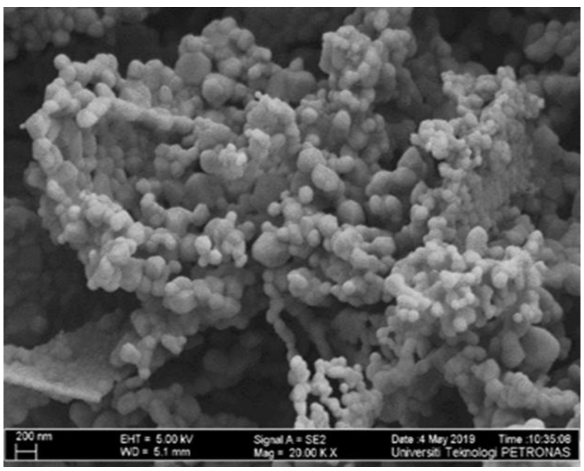


Fig. 4. FESEM image of zinc oxide nanoparticles

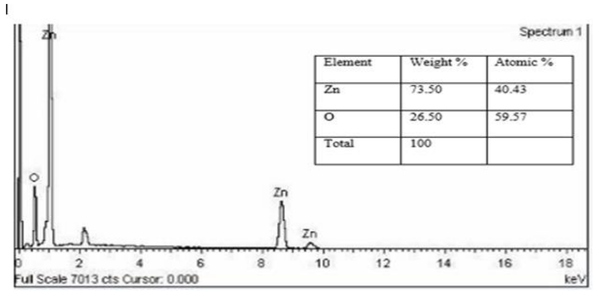


Fig.5. EDX result showing elements composition and distribution

4.2.2 Interfacial tension measurement

The potential of zinc oxide nanostructures at reducing interfacial tension at room temperature and high temperature of 95°C were investigated using force based dataphysics tensiometer. The oil-brine interfacial tension was first measured as a control before subsequent runs of nanoparticles addition on oil-brine IFT. As shown in Table 1, all measurements were repeated three times to reduce any possible error. The measured IFT value for oil and brine was 37 mN/m which is consistent which is between the reported range [19]. Table 1 presents the IFT result obtained at 25°C under different concentrations while figure 6 present the influence of concentration on IFT graphically. 0.05 wt% zinc oxide is marked as optimum concentration at which IFT reduction was achieved Therefore, this concentration was used for further test at high temperature. At this elevated thermal condition, temperature caused increase in IFT between oil and brine, from 37 to 41 mN/m [20]. However, when oil-ZnO nanofluid (0.05 wt%) were subjected to this temperature, a further decrease was observed from 27.74 mN/m to 22.87 mN/m indicative that zinc oxide exhibits pyroelectric effect [21]. This might be the cause of improvement in their abilities at reducing IFT further in the operated condition, as similar observation for pyroelectric NPs has been reported [22].

Table 2. IFT results at different concentration

Sample	Interfacial tension (mN/m) at 25°C				95°C
	1st Trial	2nd Trial	3rd Trial	Average	
Oil-Brine	33.97	38.57	38.46	37.00	
Oil-ZnO(0.1g)	34.34	34.57	34.87	34.59	
Oil-ZnO (0.3g)	32.26	32.47	32.68	32.47	
Oil-ZnO (0.5g)	27.64	27.84	27.74	27.74	22.87
Oil-ZnO(0.7g)	28.94	28.78	28.86	28.86	
Oil-ZnO(0.9g)	31.17	31.85	31.52	31.51	

5. Conclusion

In this study, the impact of nanoparticles as a suitable candidate for lowering interfacial forces is reported. The reduction in stress between solid/liquid interface is an important parameter for indicating fluid flow over the surface. Before IFT test, evaluation of dielectric nanoparticles on the stress between oil and rock was simulated at high temperature condition. The result shows that zinc oxide nanoparticles is able to perform better at high temperature. Moreso, at higher concentration under ambient condition, increase in IFT is due to agglomeration effect. Conclusively, zinc oxide nanoparticles are able to influence interfacial forces optimally at reservoir high temperature and therefore potentially enhanced displacement efficiency.

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To whom correspondence should be addressed: Yusuff Afeez, Department of Fundamental and Applied Sciences, UTP, Malaysia