International Journal of Research in Engineering and Science (IJRES)
ISSN (Online): 2320-9364, ISSN (Print): 2320-9356
www.ijres.org Volume 10 Issue 4 ǁ 2022 ǁ PP. 46-52

Experimental Investigation on Effect of Nanoparticle for Permeability Change in Enhanced Oil Recovery

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Abstract
Nanofluid flooding has been propose as the cutting-edge technology for accessing the remaining oil in the reservoir after primary and secondary production methods. It addresses the limitation of traditional enhanced oil recovery (EOR) method like high cost of chemicals and degradation of polymers and surfactants. These fluids are prepared with nanoparticles such as Zinc oxide, Aluminum oxide, silica Oxide and Magnesium oxide, etc. They have a unique property of small size and can easily access reservoir pore spaces that other substances cannot penetrate. However, they can retain in the reservoir porous media, which can lead to reduction in the permeability of the reservoir formation. Therefore, this research centered on experimenting the effect of Nanoparticle on permeability change during enhanced oil recovery at different concentrations. Core plugs were prepared from Niger Delta sand samples and nanofluids were formulated using three different nanoparticles (Aluminum oxide, Magnesium oxide and Zinc Oxide). Three different concentrations of the nanofluids were used to flood the core plugs in the laboratory with the brine (industrial salt and water) as the dispersing agent. The change in the permeability of the core plugs were analyze before and after the flooding process. The result shows that nanoparticles adsorption during flooding increases oil recovery to 17% and permeability was reduced as the concentration increases within the range of 85 to 604 md after the flooding process. Permeability change mathematical models were also develop for zinc and magnesium oxide using multiple linear regression. The model will help to checkmate the concentration of the Zinc and Magnesium oxide nanoparticle as to reduce the permeability reduction change during core flooding.

Keywords: *Aluminum Oxide, Core Flooding, Magnesium Oxide, Nanoparticle, Permeability, Regression Model. Zinc Oxide.*

Date of Submission: 28-03-2022 Date of acceptance: 09-04-2022

I. INTRODUCTION

Hydrocarbons are a key source of energy for human civilization, and are expected to remain in many decades to come. Most of oil fields have already reached or will soon reach the declining stage of production, even though a large fraction of the original oil in place (OOIP) is yet to be recovered [1]. Energy crisis and reservoirs with declining reserves even after primary and secondary oil recovery are some of the factors that led to the development of Enhanced Oil Recovery (EOR) processes. Enhanced oil recovery, also called tertiary recovery, is the extraction of crude oil from an oil field that cannot be extracted otherwise. EOR can extract 30% to 60% or more of a reservoir's oil, compared to 20% to 40% using primary and secondary recovery [2]. Fundamental studies on enhanced oil recovery (EOR) are crucial in developing technologies that enable a high recovery factor from oil reservoirs, as cheap and reliable renewable energy sources are yet unavailable. Various EOR technologies such as thermal, miscible, and chemical processes have been used and shown to boost hydrocarbon recovery.

More recently, nanotechnology have been propose as a promising EOR method since nanoparticles can penetrate the pore throat and change the reservoir properties considerably to increase oil recovery [3]. Nanofluid flooding is a chemical enhanced oil recovery process that involves injecting nanoparticles into oil reservoirs to cause oil displacement or increase productivity.

The main idea behind the application of nanoparticles in the petroleum industry concerns the inherent features of materials at the nanoscale, which ranges to very small dimensions (1-100nm). They possess unique properties such as wettability control, viscosity increase in an aqueous solution, viscosity decrease at reservoir condition, greater surface area to volume ratio, and interfacial tension reduction due to their small sizes ([3], [4]). The larger the amount of particles that are expose to surrounding materials as their surface area increases, the more materials are visible to potential reactivity. This unique attribute aids in oil recovery because their small size allows them to penetrate pores that regular EOR technologies are unable to reach. Primary and secondary oil recovery focuses on the mobile oil in the reservoir but tertiary or enhanced oil recovery aimed at the immobile oil (i.e. oil that can't be produced due to capillary and viscous forces, or what's left after conventional production), which accounts for roughly two-thirds of the original oil in place [5].

Nanofluids are created by adding nanoparticles to a base fluid in order to improve some qualities at low dispersing medium volume concentrations. Some of these nanoparticles are Aluminium oxide (Al_2O_3), Tritium oxide (TiO_2), Calcium carbonate ($CaCO_3$), Silicon oxide (SiO_3), Nickel oxide (NiO) and Zirconium oxide (ZrO_2). Researchers discovered that these tiny particles could enter into pore spaces that conventional oil recovery techniques cannot, resulting in a greater recovery [6]. Factors affecting nanofluid-flooding recovery are Concentration of the nanoparticles, Size of nanoparticles, Salinity, Temperature, Wettability of the nanoparticle, Rock grain size, Clay content, Reservoir permeability and Rate of injection [3].

Over the past years, numerous studies have shown promising results of nanomaterials application for improving hydrocarbon recovery ([7], [8], [9], [10], [11], and [12]). [7] did a research on Nanofluid coreflood experiments in Arad. He studied the flooding performance of modified nanoparticles and reported that the carbon-based fluorescent NPs increase the oil recovery factor in carbonate reservoir by more than 96%. [8] did a work on enhanced oil recovery using selected nanoparticles such as Aluminuim, Zinc, Magnesium, Iron, Zirconium, Nickel and Silicon oxides. The authors used different dispersing agent of diesel, distilled water, brine and ethanol. They reported from their experimental result that Aluminium oxide and Silicon oxide are good enhanced oil recovery agent as to compare to other nanoparticle investigated using ethanol as the dispersing agent. They concluded that oxides of magnesium and Zinc dispersed in distilled water and brine cause permeability problem, which limited the recovered oil.

[9] studied on Enhanced Heavy oil recovery using TiO_2 Nanoparticle. He reported that TiO_2 nanoparticles gave an 80 % increase in oil recovery in an oil-wet sandstone. [12] did an investigation work on the effect of Copper Oxide and Alumina nanoparticles on Enhanced oil recovery in carbonate reservoirs. The flooding process was carried out using eight limestone core samples with a salinity water as the dispersing agent. They concluded that the nanoparticles gave a best recovery at low concentration than at higher concentration.

Of recent, studies has also shown that the use of nanoparticle can reduce the permeability of reservoir formation after flooding processes ([13], [6], [14], and [15]). [13] studied the effects of injected nanofluid concentration ranging from 0.5 to 0.01 wt. percentage with an injection rate 0.1 - 4 cm3 /min and pore volume of 0.2 – 3.5 PV on permeability and porosity during nanoparticles movement through Berea sandstone. They used hydrophilic silica nanoparticle of 7 nm with Synthetic seawater of 3.0 wt. % sodium chloride. The authors examined the retention of nanoparticles during flooding experiment in three different ways, which includes; continuously increasing pressure during single-phase coreflood experiment, microscopic visualization under Scanning Electron Microscope (SEM) integrated with Energy Dispersive X-Ray Spectroscopy (EDX) as to differentiate nanoparticles with other elements, and particle measurement between influent and effluent. They reported impairment range of 5 to 88% for liquid permeability measurement and 1 to 11% for porosity.

[6] did a work on permeability alteration using silica and Alumina oxide nanoparticles for enhanced oil recovery. They conducted the experiments using core samples made with Niger Delta sand samples for both homogeneous and heterogeneous formation. The nanofluids were prepared using two different nanoparticles, with brine as the dispersing medium and different concentrations were used to flood the core sample. They concluded from their research that the use of nanoparticles increases recovery but reduced the permeability of the formation after flooding process. They also built two mathematical regression models for predicting changes in permeability for Aluima Oxide and Silica Oxide.

[14] researched on the effect of Magnesium oxide, Alumium Oxide and Silicon oxide in porous media at 45^oC and 3000 – 3500 Pisa. They reported that Aluminium oxide gave the highest recovery as to compare to other nanoparticles investigated. The authors also mentioned that increase in nanoparticle concentration increases the oil recovery but decreases the permeability of the reservoir formation after the flooding procedures. They reported that only Alumuiunm Oxide is economical at 0.2%.

1.1.1 Application of Nanotechnology

Nanotechnology application has proffered solutions to some issues ravaging the oil and gas industry ranging from petroleum exploration, drilling and completion, flow assurance problems, hydraulic fracturing and EOR. Recently nanoparticles have found application in chemical enhanced oil recovery (EOR) due to their ability to react with amphiphilic and macromolecular components of enhance oil recovery (EOR) chemicals [16]. When reacting with surfactants, polymers, and foams, respectively they form polymeric nanofluids, nano-surfactants, and nano-stabilized foam. The formed suspension (or composite materials as the case maybe) exhibit fascinating properties that form the individual components of the material. The presence of nanoparticles improves rheological behavior and prevents degradation of the polymer molecules. Nanoparticle-stabilized foams have a longer half-life. Additionally, the lamellae bubbles are thicker, hence, stable thermodynamically. Nano-surfactants have ultra-low interfacial tension (IFT) due to the irreversible adsorption of nanoparticles at the interface. Overall, during the various tests conducted in porous media, Nano-chemicals have lower adsorption hence, deemed more suitable and tagged the next revolution for the application of chemical enhance oil recovery (EOR) methods [17].

II. METHODOLOGY

2.1 Materials

The experiment involved the use of nanofluid on encapsulated plugs of unconsolidated sand (sandstone) which depicts Niger Delta formation. The materials used to achieve this purpose include encapsulated plug sample, Crude oil, Brine (the dispersing agent), Nanoparticles (Aluminium oxide, Magnesium oxide and Zinc oxide).

2.2 Laboratory Equipment Used are:

i. Canon U-tube viscometer**-** Used to measure viscosity
ii. Thermometer – Used to measure temperature
iii. Pycnometer – Used to measure density
iv. Hydrometer**-** used to measure fluid properties
v. Liquid permeability flow loop – used to determine permeability

The dispersing agent used was brine. It was prepared in the laboratory with a concentration of 30g/L. The core plug samples of 83-87cm^3 in volume was used for this experimental study. Three different samples of Nanoparticles used to perform this experiments includes; Aluminium oxide (Al_2O_3), Magnesium oxide (MgO) and Zinc oxide (ZnO). They were selected because they have been proved to be among the best nanoparticle for enhanced oil recovery. The crude oil has a specific gravity of 0.84, density of 0.87g/cm3, viscosity of 7.74cp and API gravity of 36.95. The concentration of various nanoparticles used were 5000ppm, 7000ppm and 10000ppm. Table 1 shows some of the properties of fluids used to conduct the experiment.

Table 1. Properties of the Fluids Used for the Experiment

Fluid sample	Temp (^OC)	Efflux time (sec)	Viscometer constant (150/60lb)	Density of fluid (g/cm^3)	Kinematic viscosity	Dynamic viscosity (cp)
Brine	29.00	72.00	0.03641	1.0207	2.6219	2.6761
5000ppm						
Al_2O_3 (A1)	29.00	73.00	0.03641	1.0002	2.6583	2.6588
MgO (M1)	29.00	54.00	0.03641	1.0022	1.9464	1.9507
ZnO (Z1)	29.00	42.68	0.03641	1.0020	1.5542	1.5573
7000ppm						
Al_2O_3 (A2)	29.00	105.0	0.03641	1.0008	3.8236	3.8267
MgO (M2)	29.00	60.02	0.03641	1.0032	2.1856	2.1926
ZnO (Z2)	29.00	50.56	0.03641	1.0024	1.8411	1.8455
10000ppm						
Al_2O_3 (A3)	29.00	73.12	0.03641	1.0140	4.4013	4.4629
MgO (M3)	29.00	91.00	0.03641	1.0048	3.3138	3.3297
ZnO (Z3)	29.00	53.27	0.03641	1.0032	1.9398	1.9460

2.3 Experimental Setup

The Niger-Delta unconsolidated homogenous core plug samples were prepared using four different grain sizes of 200, 400, 600, and 800 all in μm. The brine was formulated using a concentration of 30g/L of industrial salt. A lower salt concentration was used because it has been proved that lower salinity brine produces better results for oil recovery purposes. The prepared brine was used to make three different types of nanofluids, which are aluminium oxide nanofluid, magnesium oxide nanofluid and Zinc oxide nanofluid. The nanofluids were prepared using different hydrophilic Aluminium oxide, Magnesium oxide and Zinc oxide nanoparticles with a different varying concentration of 5.0g/L, 7.0g/L, and 10.0g/L. Table 1 shows the properties of all the fluids, which were properly determined after the formulation of the various nanofluids.

Before the main flooding, the core plug samples were soaked in brine for one day (24 hours) to start up the initial reservoir conditions, which is the initial saturation of the reservoir. The process is called imbibition, which is a situation in fluid flow process whereby the saturation of wetting phase increases thereby increasing the wetting fluid phase mobility. After the imbibition cycle, the flooding with oil started which is known as drainage cycle. The amount of water displaced by the oil during this process was calculated as the original oil in place (OOIP).

When the determination of initial oil in place (OOIP) is over, the main laboratory experimental work started with the primary and secondary recovery. For the primary and secondary recovery process, the flooding of the core plug was done with only brine, the flooding process continued and was terminated when the core plugs stared producing only brine. The production of only water shows that the primary and secondary recovery option for the core samples were exhausted and therefore the need for a tertiary recovery production method.

In tertiary production recovery, the three different nanofluids (aluminium oxide, magnesium oxide and Zinc oxide) was used in flooding the core plugs and was done until the production of oil by the fluid stopped. The production of only oil in the individual core samples showed the end of enhanced oil recovery (EOR) from the core samples used and the saturation of the unrecovered oil is residual oil saturation. This flooding process was carried out for all different types of nanofluids and with their various varying concentrations. The permeability of each plug samples was determined before and after flooding with nanofluid for each core to determine if the use of nanoparticles altered the permeability of the core plug.

III. RESULTS AND DISCUSSION

3.1 Relationship between Oil Recovery Factor and Nanoparticle Concentration

Figure 1. shows the tertiary recovery using different concentration of 5g/L, 7g/L and 10g/L of Aluminium Oxide, Magnesium Oxide and Zinc Oxide nanoparticles. In general, flooding with these three nanoparticles increases oil recovery factor. Aluminium Oxide nanoparticle at the concentration of 5g/L gave the highest recovery factor of 76% (Figure 1) as to compare to other recovery at that particular concentration. For the scenario of 5g/L Aluminium oxide and Magnesium Oxide flooding, the recovery was enhanced by 17% and13% respectively. The cumulative recovery at 7g/L concentrations is relatively high to compare with other concentrations due to high primary and secondary recovery (Figure 1). Generally, the recovery at 10g/L concentration is low most particularly for Magnesium Oxide and Zinc Oxide, which might be due to retention of these nanoparticles at pore volume (Figure 2). It could be seen from this study that at higher concentration, nanoparticles can cause damage to reservoir rock permeability and porosity thereby reducing the recovery factor.

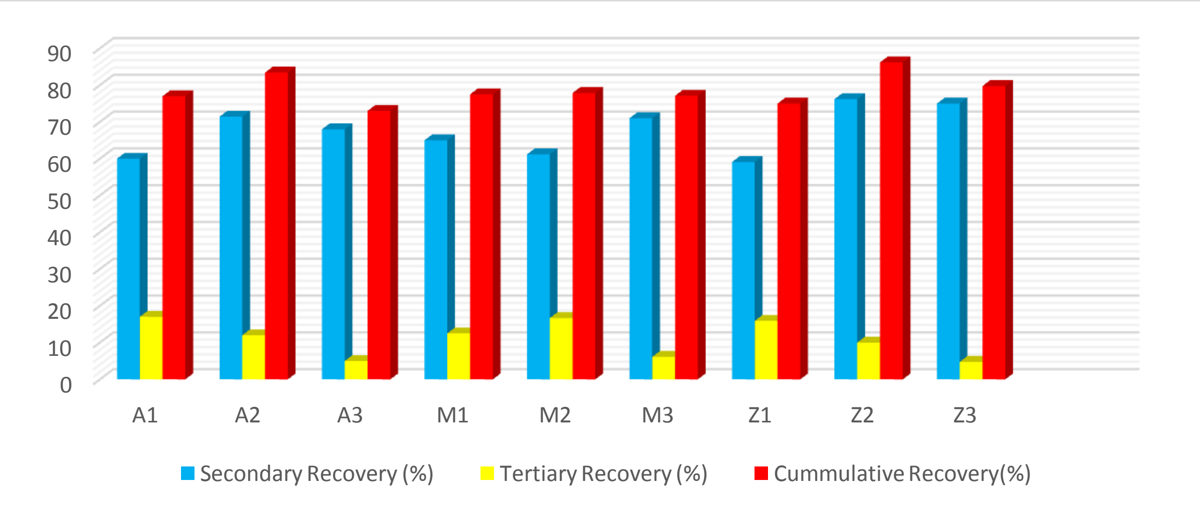

Figure 1. Percentage Recovery Plot

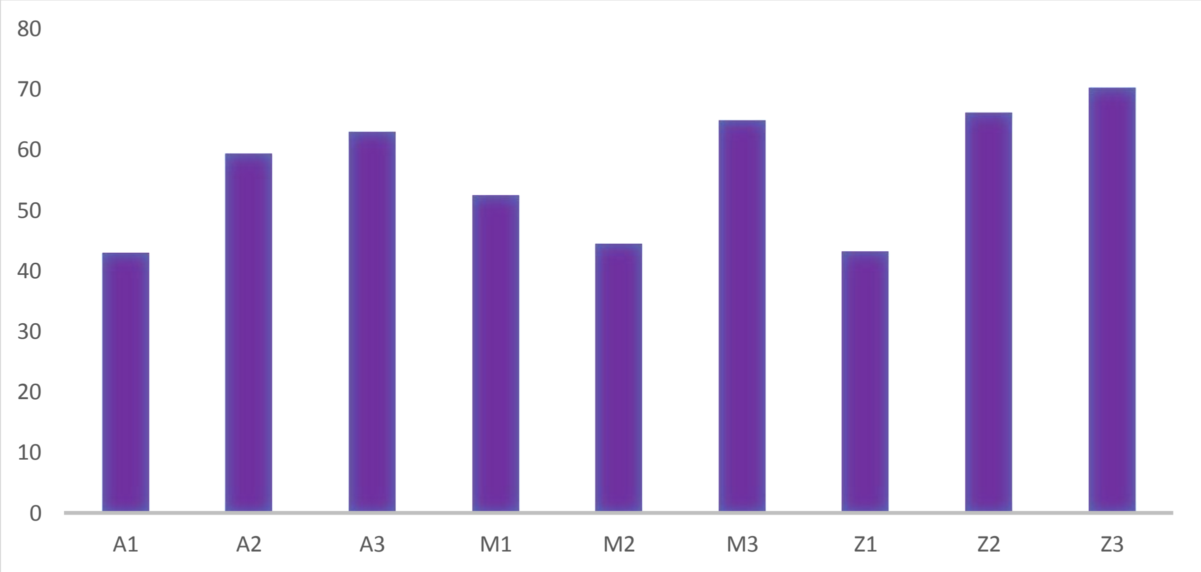

Figure 2. Change in the recovery factor Plot

3.2 Permeability Change Result

There is a significant decrease in permeability of the reservoir formation after the tertiary flooding with nanoparticles despite their good performance in enhancing oil recovery. Table 2 and Figure 3 show the changes in permeability at the different concentration of the nanofluids containing Al_2O_3, MgO and ZnO. Permeability change ranges from as low as 85.9 md to as high as 604 md. The permeability change for Aluminium oxide nanoparticle is relatively low as when compared to Magnesium oxide and Zinc oxide. Generally, the permeability change for 5g/L concentration is low and 10g/L concentration is high. This observed decrease is due the various factors like the size of the nanoparticle which can block the reservoir pore space and high flowrate. A relatively high flowrate of 0.84 cm^3/sec was obtained when flooding the core sample. It was observed that as the injection rate increases, smaller molecules of water accelerated faster than nanoparticles, assembled and block pore throat, and thus reduced oil recovery.

Table 2: Change in Permeability

Sample ID	Initial permeability	Final permeability	Change in permeability
A1	1303.07	1201.21	101.86
A2	848.93	727.79	121.14
A3	1204.01	911	293.01
M1	1057.94	972.04	85.9
M2	1268.64	1087.92	180.72
M3	1694.49	1089.84	604.65
Z1	848.93	727.79	121.14
Z2	1243.59	888.7	354.89
Z3	1303.07	931.21	371.86

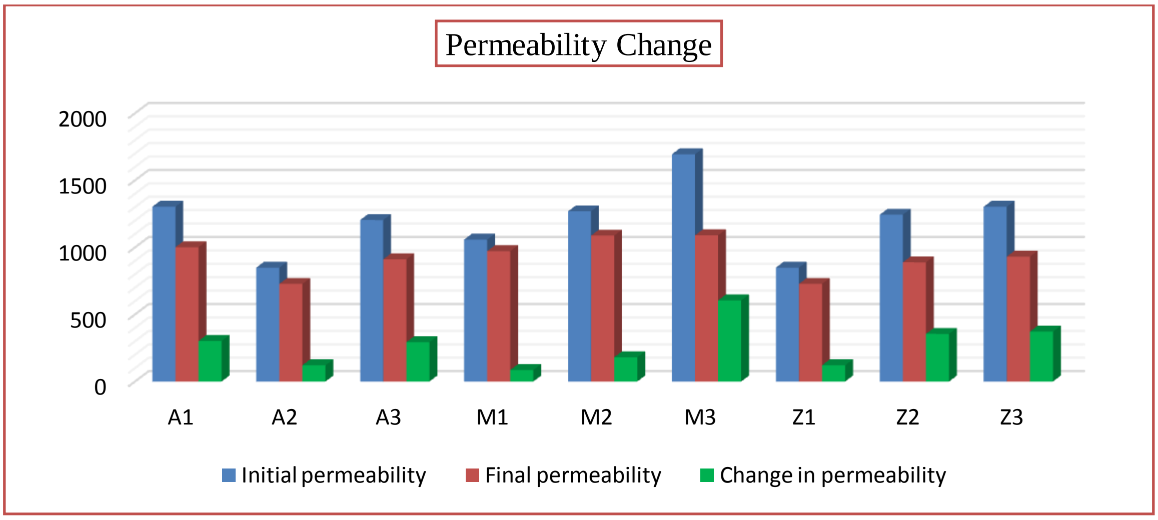

Figure 3. Permeability Alteration for Different Concentration Plot

3.3 Zinc and Magnesium Oxide change in Permeability Model

The experimental data gotten from this work was used to develop Zinc and Magnesium Oxide change in permeability models using multiple linear Regression method (Equations 1 to 3). The models depend on the concentration of the nanoparticles and the initial permeability of the reservoir formation. It helps to determine the extent of permeability alteration due to different concentrations of Zinc and Magnesium nanoparticles.

$$\Delta K = K_i - K_f \quad (1)$$

Where:

ΔK = change in permeability (permeability)

K_i = Permeability before nanofluid flooding

K_f = Permeability after nanofluid flooding

For Magnesium oxide nanoparticle;

$$K_f = 0.129371503 \; K_i + 0.017724285 \; N + 746.5512877 \quad (2)$$

For Zinc oxide nanofluid:

$$K_f = -0.175559279 \; K_i - 0.099250868 \; N + 2099.53244 \quad (3)$$

Where N is the concentration of nanofluid for both Equations 2 and 3.

Assumptions for the models; the reservoir is isentropic, the reservoir fluid is incompressible, the reservoir is saturated with a single fluid, there is no interaction between the reservoir rock and nano-fluid and the core sample is homogeneous.

The statistical parameters of correlation of coefficient (R) and absolute percentage error (ARE) were used to analyse the performance of the new models. Both models gave coefficient of correlation of 1 and absolute percentage error of 0 respectively.

IV. CONCLUSION

Three types of nanomaterials at three different concentrations of 5g/L, 7g/L and 10g/L were used to investigate their effects on enhanced oil recovery and permeability. Based on the experimental results, all the nanoparticles studied increased oil recovery but Aluminium Oxide gave the highest recovery of 17% with Zinc Oxide the lowest. The 5g/L concentration averagely gave the highest recovery than 7g/L and 10g/L while 10g/L concentration averagely has the highest permeability reduction. A simple regression models for permeability change were developed for Magnesium and Zinc Oxide nanoparticle. Nanofluid concentration in the range of 5 to 7g/L might be favourable for enhanced oil recovery core flood experiment to minimize possibility of formation damage due to permeability reduction

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