

Permeability Alteration Due to Nanoparticles Retention in the Porous Media during Nanotechnology Assisted Enhanced Oil Recovery Process

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Abstract– Nanofluid flooding has been proposed to be a ground breaking technique for the recovery of the inaccessible oil in place in the reservoir after primary and secondary production methods. It is said to have the ability to overcome problems faced by traditional EOR methods. These fluids are prepared with the use of nanoparticles, which due to their small sizes possess some unique properties and can easily pass-through reservoir pore spaces that other substances cannot reach, resulting in the adsorption of the nanoparticles in the reservoir, leading to positive changes in reservoir microscopic properties and a corresponding increase in oil recovery. However, these particles can also be retained in the reservoir porous media, and can lead to a reduction in the permeability of the reservoir. Experiments were conducted using core samples made with Niger Delta sand samples. Nanofluids made using two different nanoparticles, with brine as the dispersing medium and different concentrations were used to flood the core plugs. The results from the experiments were analyzed using charts to check the effectiveness of the process. Also, the differences in the permeability of the plugs before and after flooding were compared. The results show that nanofluids adsorption during flooding causes an increase in recovery but also reduced the permeability significantly after the flooding process. Two mathematical models were created using multiple linear regressions for each type of nanoparticle used. The models predicted permeability alteration effectively with a correlation coefficient of one. This shows that the models can be used to predict accurately the changes in permeability during a nanofluid flooding process after computing the right parameters.

Keywords– Enhanced Oil Recovery, nanoparticles, nanotechnology, permeability alteration, porous media

1 INTRODUCTION

Nanotechnology for enhanced oil recovery has been tested by several parties and has proven to have the ability to recover the inaccessible oil reserves that conventional recovery methods cannot reach. Comparison of the exceptional properties of nanoparticles to the bulk material counterpart shows that nanoparticles show different properties and superior behavior [12], hence their use for oil recovery.

The main idea behind the application of nanomaterials in the petroleum upstream industry concerns the inherent features of materials at the nanoscale. They possess unique properties due to their small sizes and greater surface area to volume ratio.

As the surface area of particles increase, the greater the amount of particles that comes in contact with surrounding materials resulting in more materials being exposed for potential reaction [28]. This is one unique property of nanomaterials which helps in improving oil recovery as these small sizes gives them the ability to penetrate some pores that traditional EOR methods cannot reach.

The recovery efficiency of conventional secondary oil recovery methods usually depends on the mechanism of pressure maintenance, hence their inability to access certain reserves. To solve this problem, EOR methods involves the injection of fluids into the reservoir which interacts with the reservoir rock and fluid systems causing changes to certain rock and fluid properties, thereby increasing recovery. Studies conducted on the application of nanotechnology to enhance oil recovery shows that the injection of nanoparticles into oil reservoirs causes trapped oil in pores to be released by reducing the interfacial tension between the oil and water, the wettability alteration of the porous media and the modification of flow character which ultimately increases oil recovery [1].

The world's increasing energy demand has created the need for more recovery of crude oil from oil fields. Primary and secondary production methods have been able to recover only about a third of the original oil in place (OOIP), leaving a substantial amount of oil underground. This challenge has brought about the need for advanced tertiary recovery methods also known as Enhanced Oil Recovery (EOR), which tends to

bridge the gap between the increasing global energy demand and the unsatisfactory recoveries from conventional (primary and secondary) production methods.

While primary and secondary recovery targets the mobile oil in the reservoir, tertiary or enhanced oil recovery targets the immobile oil (i.e. oil that cannot be produced due to capillary and viscous force or what is left after the conventional production method), that is about two-thirds of the oil in place [2].

The interest of the application of nanotechnology in the petroleum industry has been on a steady rise in recent years due to its successful application in other industries and studies have shown that they offer promise for future enhanced oil recovery processes.

Compared with conventional chemical flooding, nanofluid flooding has several advantages. Nanofluid flooding is defined as a chemical enhanced oil recovery technique whereby nanomaterials are injected into oil reservoirs to effect oil displacement or to improve productivity [1]. The nanofluids are made by the addition of nanoparticles to a base fluid for the improvement of some properties at low volume concentrations of the dispersing medium [7].

Although the oil displacement mechanisms with nanoparticles has not yet been clearly understood, recent studies have shown that they offer good application for unlocking the remaining oil stocked in the pores of the reservoir. Several authors have started to investigate on a lab scale, the usefulness of the nanofluids for improving oil recovery and they have observed that nanoparticles enable a higher recovery of about 80-90% of the oil in place [14].

Particles have shown better properties when reduced to nano-scale, the enhanced properties brought about as a result of the reduced size of the particles includes the large surface area-to-volume ratio which causes enhanced activity due to an increase in contact area, and the chemically modified surfaces leading to wettability alteration at nanoscale.

Researchers have observed that these tiny particles can penetrate into the pore spaces that conventional oil recovery techniques are not able to do and therefore resulted in higher recovery [23]. These studies also showed that nanoparticles can be tailored to alter reservoir properties such as wettability, interfacial tension reduction and control of fines migration.

By changing the formation wettability from oil-wet towards a more water-wet condition, capillary force will decrease and the mobility of the oil phase will be enhanced thereby causing a higher recovery.

However, these particles are retained and sometimes trapped in the pore spaces of the reservoir which causes the plugging of the pores, thereby reducing the pore size which will eventually cause a reduction in the reservoir permeability. This may be caused by the particle size being larger than the pore channels or as a result of log-jamming, the plugging of pore channels that are larger than the nanoparticles resulting in nanoparticles accumulation, leading to the blockage of the underground pore networks. This may also result in formation damage; hence, the need to study the effect of nanofluid flooding on reservoir permeability.

1.1 Types of Nanoparticles Used For EOR

For enhanced oil recovery, the choice of nanoparticles to be used is divided into three different categories, namely: Metal oxide nanoparticles, Organic nanoparticles and Inorganic nanoparticles [4].

1.1.1 Metal oxide nanoparticles

The commonly used nanoparticles in this category include aluminum oxide, copper oxide, Iron oxide, tin oxide and zinc oxide with aluminum oxide being the most frequently used because of its ability to reduce the interfacial tension of oil and brine, and the oil viscosity [4]. Experiments conducted using aluminum oxide, shows that it gives a higher recovery when brine is used as the dispersing agent and that it would be the best

option for heavy oil reservoirs [3]. Also, aluminum oxide has provided better results for EOR application when used in combination with silica (an inorganic nanoparticle).

1.1.2 Inorganic nanoparticles

The most commonly used nanoparticles in this category is silica (SiO_2). Silicon dioxide is one of the most abundant compounds on earth, hence why it is the most frequently used and it is a very cost effective nanoparticle. According to Shidong et al. [25], silica is considered very suitable for water-wet sandstone reservoirs because it can easily alter the wettability to intermediate-wet and it also reduces the interfacial tension of such reservoirs. Furthermore, it has also proven to be very stable in high temperature conditions. Experiments conducted using silica nanoparticles with Berea sandstone and brine showed that silica improved the recovery with about 48.7% by changing the wettability from water-wet to intermediate-wet sandstone. Hydrophilic silica nanoparticles can be absorbed on pore walls of sandstone reservoirs giving rise to pore blockage and the wettability of the reservoir surfaces [5]. Other inorganic nanoparticles include zeolite and polymer nanoparticles.

1.2 Methods of Applying Nanoparticles

1.2.1 Nanofluids

Since nanoparticles cannot be injected directly into the reservoir, they are injected in the form of fluids called nanofluids. Nanofluids are created by the addition of nanoparticles to a base fluid [27]. The base fluid could be water, oil or gas. Due to the inability of nanoparticles to remain in suspension, a stabilizing agent is added, especially to hydrophobic nanoparticles to prevent the clustering of the particles. Nanofluids possess the unique properties of the nanoparticles used in making them and offer great potential for many applications. A number of factors affect the properties of a nanofluid [20]. These factors include:

1. The type of nanoparticles used in making them.
2. The concentration of the nanoparticles.
3. The particle size and morphology of the suspended particles.
4. The distribution of the dispersed nanoparticles.

There's no denying the fact that nanofluids has great potential for improving recovery but the problem of stability is of great importance. Nano-fluids consist of two phases – liquid and solid phases – and with the two phase's system come the problem of stability [18]. For better results, there is a need to achieve the desired stability of nanofluids before their use. Although a two-step method which involves the production of nanomaterials as dry powders first before dispersing them in a fluid, and then using surfactants to stabilize the fluid is the most economic and widely used method; researchers have discovered that it isn't a very effective method in preparing stable nanofluids [12], hence the need to develop new and improved methods for its production.

The introduction of the one step method has been a welcoming idea. This method helps to reduce the agglomeration of the nanoparticles caused by the high surface area of the nanoparticles and in the process increases the fluid stability. The limitation of this method is its very high cost and that it cannot be used to produce nanofluids in large scale [13]. More methods are being developed to fix this problem.

1.2.2 Nanocatalysts

Generally, catalysts are materials that increase the rate of chemical reactions without getting themselves used up in the process. Therefore, nano-catalysts can be defined as nanoparticles or nanomaterials that can be used to improve chemical reactions. They are usually metallic nanoparticles. Compared to other catalysts, they have some advantages because of the unique properties of their reduced size; the large surface area to volume ratio of nano-catalysts increases the number of atoms on their surfaces causing more reactivity when compared to bulk-sized catalysts.

They are used to improve the recovery in heavy oil reservoirs by reducing the viscosity of heavy oils like bitumen, converting them to lighter products by some chemical reactions. The reactions are called aquathermolysis [28].

1.2.3 Nanoemulsions

Nanoemulsions are emulsions stabilized using nanoparticles. They are more stable than conventional emulsions stabilized by surfactants or colloid solids, hence their ability to overcome the challenges encountered while using these conventional emulsions. They can withstand harsh reservoir conditions and their high viscosity is very effective in controlling mobility ratio, which explains their use in increasing oil recovery [12].

1.3 Factors That Affect Oil Recovery When Using Nanoparticles

1.3.1 The type of nanoparticles used

The type of nanoparticles used for flooding influences oil recovery factor and the selection of appropriate nanoparticles for typical reservoir conditions is of utmost importance. While different nanoparticles can be used to improve oil recovery, some show more promise than others. Many literatures, in their review of the application of nanotechnology in enhanced oil recovery, observed that silicon oxide nanoparticles showed more tendencies to improve oil recovery than iron oxide nanoparticles and that aluminum oxide had more tendencies to enhance oil recovery than the both of them [15].

Experiments conducted using three different types of nanoparticles – Al_2O_3 , TiO_2 and SiO_2 – to determine their ability to displace oil from an intermediate-wet sandstone core sample showed that, at the same injection conditions, Al_2O_3 , TiO_2 and SiO_2 increased the oil recovery to 52.6%, 50.9% and 48.7% respectively, after using brine for flooding tests conducted initially to establish the initial recovery before the use of nanoparticles at 47.3% [19]. These results show that the type of nanoparticle used for an EOR process would indeed affect the recovery of that process.

1.3.2 Concentration of the Nanoparticle

The concentration of the nanoparticles used is the most important factor to consider when conducting a nanoparticle assisted EOR process [1]. Flooding experiments were conducted using different concentrations of hydrophilic silica nanoparticles and their results showed that an increase in the concentration of silica nanoparticles used caused an increase in oil recovery [7].

Other researchers have also discovered that for a very effective EOR process, an optimal nanoparticles concentration has to be determined, because a higher concentration after that point may have no effect on recovery other than increasing the cost of the EOR process and in some cases, it may even reduce recovery [21].

Other experiments were also conducted to determine the effect of increasing the concentration of nanoparticles on recovery, using Iranian crude oil samples, nanosilica particles and real carbonate reservoir core samples. The results of the experiments showed that an increase in the concentration of the nanosilica particles led to a decrease in residual oil saturation which means that there was a steady increase in oil recovered as concentration increased [8].

2. MATERIALS

The materials used to conduct the experiments for this work included:

1. Brine (used as the dispersing agent);
2. Sand;
3. Crude oil; and
4. Nanoparticles.

The laboratory apparatus used included:

1. Viscometer
2. Pyrometer
3. Hydrometer – used to measure fluid properties
4. A flooding set up for the flooding process
5. A liquid permeameter flow loop – used to determine permeability.

The brine was laboratory prepared with a concentration of 30g/L. Encapsulated plug samples of 83-87cm³ in volume were used. Two types of nanoparticles were used to perform the experiments – aluminum oxide (Al₂O₃) and silicon dioxide (SiO₂). They were chosen because they have been proved to be the best for enhanced oil recovery purposes.

Table 1: Some properties of fluids used to conduct the experiment.

Fluid samples	Temp (°c)	Efflux Time (sec)	Viscometer constant 150/601B	Density of fluid (g/cm ³)	Kinematic viscosity	Dynamic viscosity (CP)
Brine	29	24.00	0.03641492	1.020	0.8740	0.9367110052
Crude oil	29	13.00	0.03641492	0.849	0.4734	0.9370770509
5000ppm						
SiO ₂ NF	29	25.50	0.03641492	1.0190	0.9286	0.9437058777
Al ₂ O ₃ NF	29	25.00	0.03641492	1.0186	0.9037	0.985537598
7000ppm						
SiO ₂ NF	29	25.80	0.03641492	1.0186	0.90327	0.9622574616
Al ₂ O ₃ NF	29	26.20	0.03641492	1.0186	0.9471	1.083292955
10000ppm						
SiO ₂ NF	29	26.60	0.03641492	1.0162	0.9616	0.9959631378
Al ₂ O ₃ NF	29	26.89	0.03641492	1.0196	0.9721	1.083027676

3. EXPERIMENTAL PROCEDURES

Encapsulated core plug samples of the unconsolidated Niger Delta sand formation samples were prepared using four different grain sizes (212 μm , 425 μm , 615 μm and 800 μm). These grain sizes were used to prepare both homogeneous and heterogeneous core samples for good comparison of results found in different types of formations.

The brine was also prepared using a concentration of 30g/L of salt. A lower salt concentration was used because it has been proved that lower salinity brine produces better results for recovery purposes. The already prepared brine was also used to make two different types of nanofluids (alumina NF and silica NF) using hydrophilic silica and alumina nanoparticles of varying concentration of nanoparticles (5g/L, 7g/L and 10g/L) for both fluids. After which the properties of all fluids were adequately determined.

Prior to the main experiment (flooding), the core plug samples were soaked in brine for 24 hours (imbibition cycle) to initiate initial reservoir conditions, i.e., the initial saturation of the reservoir formation completely with only brine. After the core samples were completely saturated with brine, the flooding of the plugs with oil commenced (drainage cycle); the amount of water displaced by the oil during this process was computed as the oil originally in place (OOIP).

After determining the initial oil in place, the main experiment commenced with the primary and secondary recovery determined by flooding the core plugs with only brine, this process was performed until the core plugs stopped producing oil and started producing only brine. The production of only water shows that the primary and secondary production option for the core samples were exhausted and hence the need for a tertiary method of production. When the flooding with brine was concluded, flooding with the nanofluids (NF) commenced (EOR) and was done until the production of oil by the fluid stopped. This marked the end of recovery from the individual core samples and the saturation of the unrecovered oil is termed the residual oil saturation. This process is conducted for all types and concentration of nanofluids.

The permeability of each plug sample was determined before flooding and after flooding with nanofluid for each core to determine if the use of nanoparticles altered the permeability of the core plug in any way.

4. RESULTS AND DISCUSSIONS

Table 2 below shows that there was actually an increase in oil recovery with the use of nanoparticles in the form of nanofluid. With samples having single letters representing homogeneous samples and those with multiple letters representing heterogeneous samples, it can be seen that nanofluid (using SiO_2 and Al_2O_3) increased the recovery with an additional average of 13.2 %, whereas brine could not. Therefore, the cumulative percentage of oil recovered was increased by a range of 55% - 75%. Alumina nanofluid recovered more for the homogeneous grain size samples, depicting a homogenous reservoir (A2 and D2) compared to flooding with SiO_2 nanofluid (A1 and D1). While the Silica nanofluid proved to be more effective for heterogeneous grain size samples, depicting a heterogeneous reservoir (CD1, ABC1 and ABCD1) compared to when flooding with Al_2O_3 nanofluid (CD2, ABC2 and ABCD2).

Table 2: Experimental results to determine production for crude oil in the different samples

Sample	Volume of oil in plug (OIIP) (cm ³)	Secondary Recovery		Tertiary Recovery		
		H ₂ O Prod (ml)	Oil Prod (ml)	Silica NF (ml)	Alumina NF (ml)	Residual Oil (ml)
A1	13.5	2.5	8.0	1.5	-	4.0
A2	13.2	2.1	8.1	-	1.6	3.5
D1	16.5	3.2	8.8	2.0	-	5.7
D2	15.9	3.0	9.0	-	2.5	4.4
CD1	18.0	4.1	9.5	2.2	-	6.3
CD2	20.0	1.5	11.5	-	1.8	6.7
ABC1	12.2	2.0	4.0	3.0	-	5.2
ABC2	20.0	2.7	10.0	-	1.5	8.5
ABCD1	13.5	2.5	7.0	2.5	-	4.0
ABCD2	17.5	3.1	9.5	-	1.8	5.7

In general, the experiment showed that using nanoparticles for EOR increases oil recovery. Since EOR methods have been proved to increase oil recovery by 10 – 20% in addition to the 50% oil recovered after primary and secondary recovery. Figure 1 below gives a clearer picture of the results.

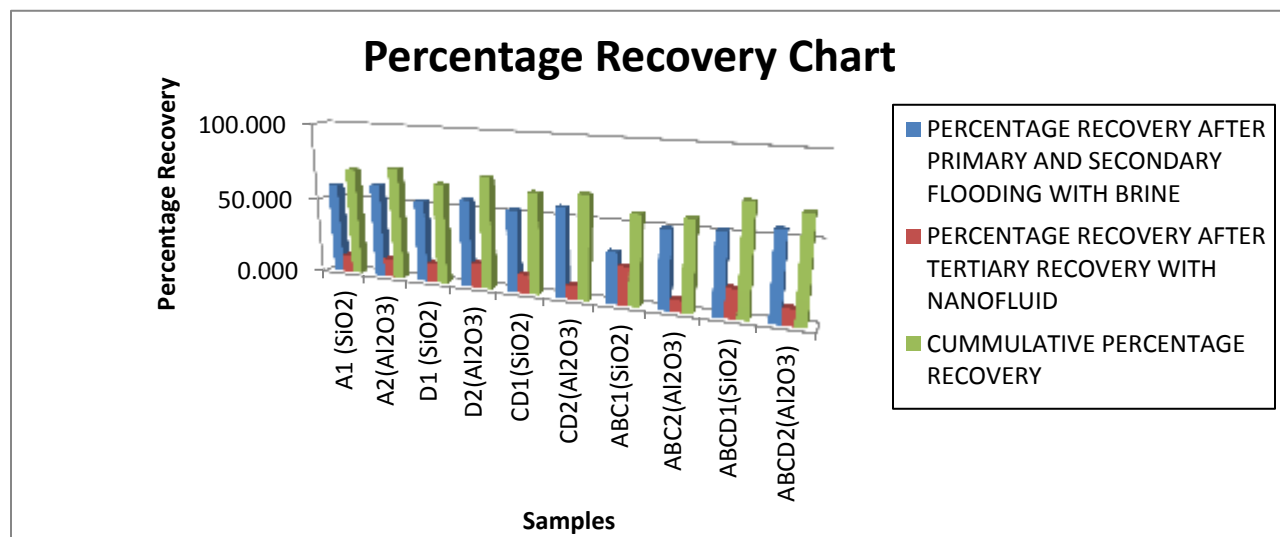


Figure 1: Percentage recovery chart

Although the use of nanoparticles for enhancing oil recovery showed good results, determination of permeability of the core plug samples after the tertiary recovery process showed a significant decrease in the recovery of the unconsolidated formation core samples, with different nanofluids, and a difference in concentration showing different results. At lower concentrations (5000-7000 ppm), Silica-nanofluid flooding resulted in higher permeability alterations in comparison to flooding with Alumina-nanofluid. Permeability alteration in the range of 200 mD – 600 mD was recorded. In contrast, at higher concentrations (10,000ppm) Alumina-nanofluid flooding resulted in a higher permeability alteration in heterogeneous samples compared to flooding with Silica-nanofluid.

This reduction in the permeability of the formation may be due to the retention of the nanoparticles used in making the fluid in the pores of the reservoir and its accumulation would cause the blockage of the pore network of the reservoir over time.

Table 3: Determination of Permeability Alteration

Sample	K_f (mD)	K_i (mD)	$\Delta K = K_i - K_f$ (mD)
A1	381.7132	1062.0716	680.3584
A2	379.6482	617.4552	237.8070
D1	302.7452	474.1087	171.3635
D2	276.7154	640.4509	363.7355
CD1	415.5944	971.5848	555.9904
CD2	246.5297	959.2239	712.6942
ABC1	455.9208	712.9846	257.0638
ABC2	280.1683	382.1586	101.9903
ABCD1	666.1171	1093.9796	427.8625
ABCD2	312.1834	1107.8512	795.6678

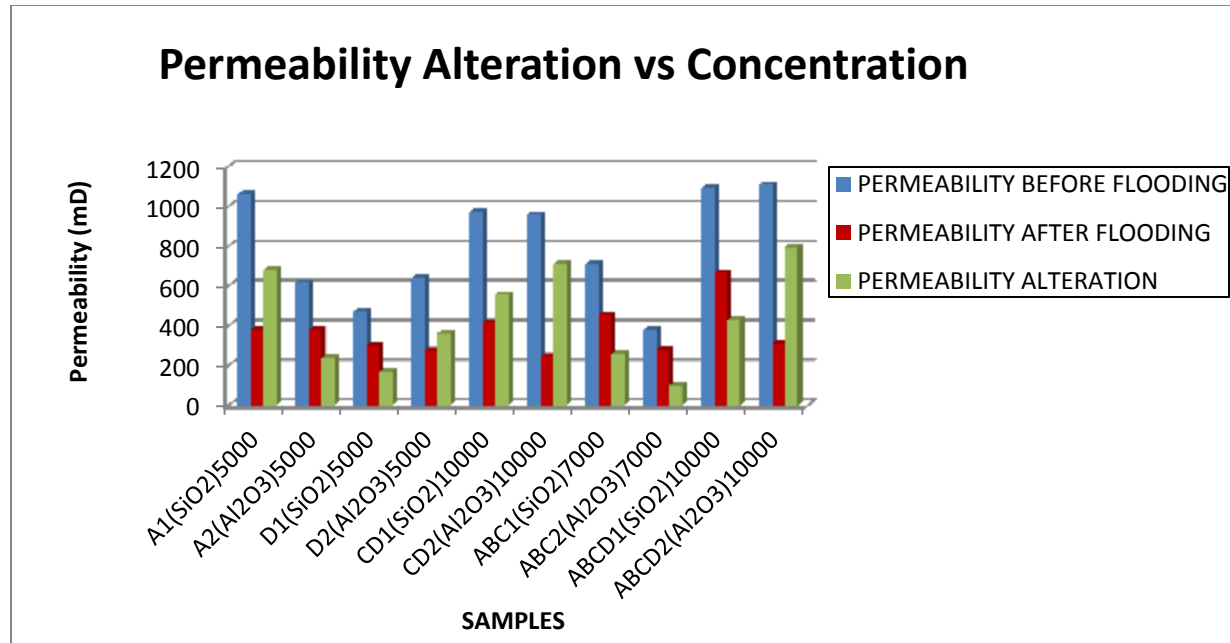


Figure 2: Permeability values vs. concentration of nanofluid and samples

Multiple Linear Regression (MLR) analysis was used to create a mathematical model that could be used to determine the permeability alteration owing to nanoparticles retention on a reservoir formation if certain rock and fluid properties are known. Two correlations were created using the experimental results computed above for silica-nanofluid and alumina-nanofluid each. The correlation forgoes the need for experiments to determine how the use of a nanofluid would affect the permeability of the reservoir. The equation for permeability alteration is given as:

$$\Delta K = K_{\text{before}} - K_{\text{after}}$$

Where:

ΔK = change in permeability (permeability alteration)

K_{before} = permeability before nanofluid flooding

K_{after} = permeability after nanofluid flooding.

Note: Permeability here is measured in Darcy (D).

The permeability after flooding is determined using the following correlations:

For Alumina-nanofluid:

$$K_{\text{after}} = 0.044592241 K_{\text{before}} - 20.47307744A - 11.94346668\phi - 0.001317012N + 920.1257941$$

Where:

A = area of the core plug, cm²

Φ = porosity, %

N = concentration of the nanoparticle, ppm.

The model for **silica-nanofluid**:

$$K_{\text{after}} = 2.463224572 K_{\text{before}} + 407.9970754L + 163.320432\phi - 0.25702484N - 9203.330871$$

All the unknowns are the same with that for alumina and L = length of core plug, cm.

The assumptions for the models are:

1. The reservoir is isentropic.
2. The reservoir fluid is incompressible.
3. The reservoir is 100% saturated with a single fluid.
4. No interaction between the reservoir rock and nano-fluids.

The correlations were used to compute the permeability values after the flooding process and the Average Absolute Percentage Error (AAPE), Root Mean Square Error (RMSE) and correlation coefficient (R) of the models in comparison with the experimental data were determined. Table 4 below gives a summary of the results obtained.

Table 4: Statistical Error Analysis

Type of Nanofluid	AAPE	RMSE	R
Silica	2.8254×10^{-13}	1.1677×10^{-12}	1
Alumina	1.8911×10^{-14}	6.7258×10^{-14}	1

From table 4 above, it can clearly be seen that the two models have a correlation coefficient of 1 and very small values (negligible) of Average Absolute Percentage Error and Root Mean Square Error. Thus, the models are very efficient in determine the permeability alteration due to nanoparticles retention in the porous media during nanotechnology assisted enhanced oil recovery.

5. CONCLUSION

The following conclusions were made after this study:

1. The use of nanoparticles for enhanced oil recovery is effective in increasing the recovery of oil from the reservoir.
2. The type of nanoparticle used has an effect in the amount of oil recovered from the reservoir.
3. The retention of the nanoparticles in the pores of the reservoir would cause a reduction in the permeability of the reservoir.
4. Different nanoparticles affect the reservoir permeability in varying degrees.
5. The mathematical model created, having a correlation coefficient of 1 and standard error of 0 can be used effectively to calculate the effect of the particle on the permeability of the reservoir without going through a tedious experimental process.

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