ASSESSMENT OF MAGNESIUM OXIDE (MGO) AND SILICA DIOXIDE (SIO_2) NANO-FLUIDS ADDITION WITH SURFACTANTS AS NEW TECHNIQUE TO CONTROL FINES MIGRATION

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ABSTRACT

Formation damage in oil reservoirs because of fines migration is among the main contributors to productivity decline. Conventionally, chemical methods such as complex organic polymers are used to prevent fines migration by stabilizing or maintain the fines particles in aggregated form. However, most of the chemical fines stabilizer is not robust, which functionalize based on the composition of exchangeable cations in clay minerals, temporary stabilization and are not environmentally friendly. In this research work, qualitative methods were developed to investigate the potential of Magnesium Oxide (MgO) and Silica Dioxide (SiO_2) Nano-Fluids addition with surfactants (Triton X-100 and Tween 80) as a new technique to control fines migration. Silicon Dioxide nanoparticles used in this experiment are in the form of colloidal silica with spherically sized at 10-15 nm then stabilized electrostatically to allow the particles to stay suspended in the solution. To measure the Nano-Fluids and fines solution stability, the minerals (kaolinite, bentonite and illite-smectite) crushed and suspended in 100 mL distilled water with 0.20%, 0.50%, 1.50% and 3% concentration, respectively. The suspended fines solutions left for 24 hours. Solution stability measured using the Zetasizer Nano ZSP by recording their turbidity (NTU) and zeta potential (mV). The results from the assessment shown that bentonite and kaolinite fines are more susceptible to migration in sandstone reservoirs due to their high colloidal stability from their zeta potential measurements and their morphologies. It can also be concluded that Silicon Dioxide (SiO_2) is more stable when these nanoparticles are suspended in de-ionized water only, which gives zeta potential values of -26.10 mV. The addition of surfactants (Triton X-100 and Tween 80) has also shown significant results on protected fines migration. Triton X-100 and Tween 80 increased Silicon Dioxide (SiO_2) Nano-fluid zeta potential value at -30.6 mV, which is beneficial for the Nano-Fluid to travel deeper into the formation to treat problematic areas of fines migration.

Keywords: *MgO and SiO_2 nano-fluid, zeta potential, fines migration, surfactant, nanoparticles*

INTRODUCTION

Fines migration is of prime importance to ensure efficient production of the well. Formation damage due to fines migration is more severe in near-wellbore areas; it reduces the productivity of oil and gas reservoirs and affects the economics of many operations in oil/gas fields [1]. Fines migration is the major formation damage mechanism, especially to sandstone formations. This study begins by thoroughly weighing the effectiveness of current options and technologies. The chosen objectives will focus on the determination of optimum Nano-Fluid (NF) concentration and zeta potential measurements between fines and Nano-Fluids. Today nanotechnology has been extensively applied in the oil industry, such as corrosion prevention, enhancement of oil recovery, improvement of cement quality and drilling operations [2]-[8].

Recently, nanoparticles have been used to control fines migration. Huang et al. [9] found that the proppant particle covered by nano-fluid could reduce fines migration significantly in filling bed. Ahmadi et al. [10] investigated the effects of SiO_2, MgO and Al_2O_3 nanoparticles on fines migration [10]-[12]. Ahmadi et al. [10] investigated the effect of base fluid salinity of nano-fluid on the ability of nanoparticles to control fines migration in porous media and proposed a mathematical model to simulate fines migration in porous media after the treatment by nano-fluid [13]-[15]. Assef et al. [16] investigated the effect of MgO nanoparticles on the control of fines particles. In this study, we assess the quantify surface forces of fines and Nano-Fluids (NF), Magnesium Oxide (MgO) and Silica Dioxide (SiO_2) through zeta potential and to investigate the best concentration of both Nano-Fluids (NF) and compare them based on their fines attachment effectiveness, especially in Malaysian oil and gas field.

BACKGROUND

Being able to produce hydrocarbons with minimal hindrance has always been the goal of developing a profitable field. To do so, one must consider the formation conditions and what damages might be caused by external factors, one of which is fines migration. It poses to be one of the more complicated cases because fines cannot be extracted to surface to be investigated conveniently due to their sizes. A reduction will typically follow the migration of fines in permeability because of its trapping mechanism after movement. Generally, the three steps of how fines contribute to formation damage are: fines detach themselves from the formation, fines break apart and travel in the pores and fines trapped and held in place (could lead to bridging, ionic attraction or wettability effects). Usually, the behaviour of fines is being observed through production curve analysis, core flooding or backflow test so that it can be evaluated with its water sensitivity concerns and critical velocity.

Scientists are determined to understand the factors contributing to this reduced permeability. According to Sakar and Sharma [17], the permeability decline has been drastic due to this problem. The results of their investigation give a permeability ratio (retained permeability to initial permeability) of 1/1000 after allowing dispersed fines to move in single-phase flows [17]. These fines move approximately less than 0.2 inches (0.5 cm) before being trapped at pore throats. Similarly, the two-phase flow has a permeability reduction of 1/50, showing the effect of the residual oil phase. Do note that during their experiment, they found that these fines released depend on the fluids saturating the core, thus brings us to conclude that the fines migration mechanism will differ from one type of reservoir rock to another [1].

The advancement of this issue has motivated researchers to understand the environment suspending these fines in the reservoir. Two of the parameters that have been studied in vast variations are salinity and pH, where both parameters characterize ionic conditions of the fluid. Once the colloidal conditions of the fluid changes, it will encourage the in-situ release of naturally existing fines (a large amount of it being clays). The change in salinity from high to low of the permeating fluid causes a massive reduction in permeability, resulting in a condition called water sensitivity (water shock). At high pH, the clay (fines) dispersion increases, becoming detrimental to formation permeability [2]. Other than that, since the movement in the reservoir is governed by flow, having high flow rates introduces a drag force on the fine particle, which is then made worst depending on its magnitude, oil viscosity and gas turbulence. At times, the problem might lie in the design of equipment, i.e., improper choke sizing, where the flow rate is not constant throughout the production period. As long as the flow rate is more than its critical velocity of fine particles, fines migration will be an issue in that reservoir. The factors mentioned above illustrated in Figure 1.

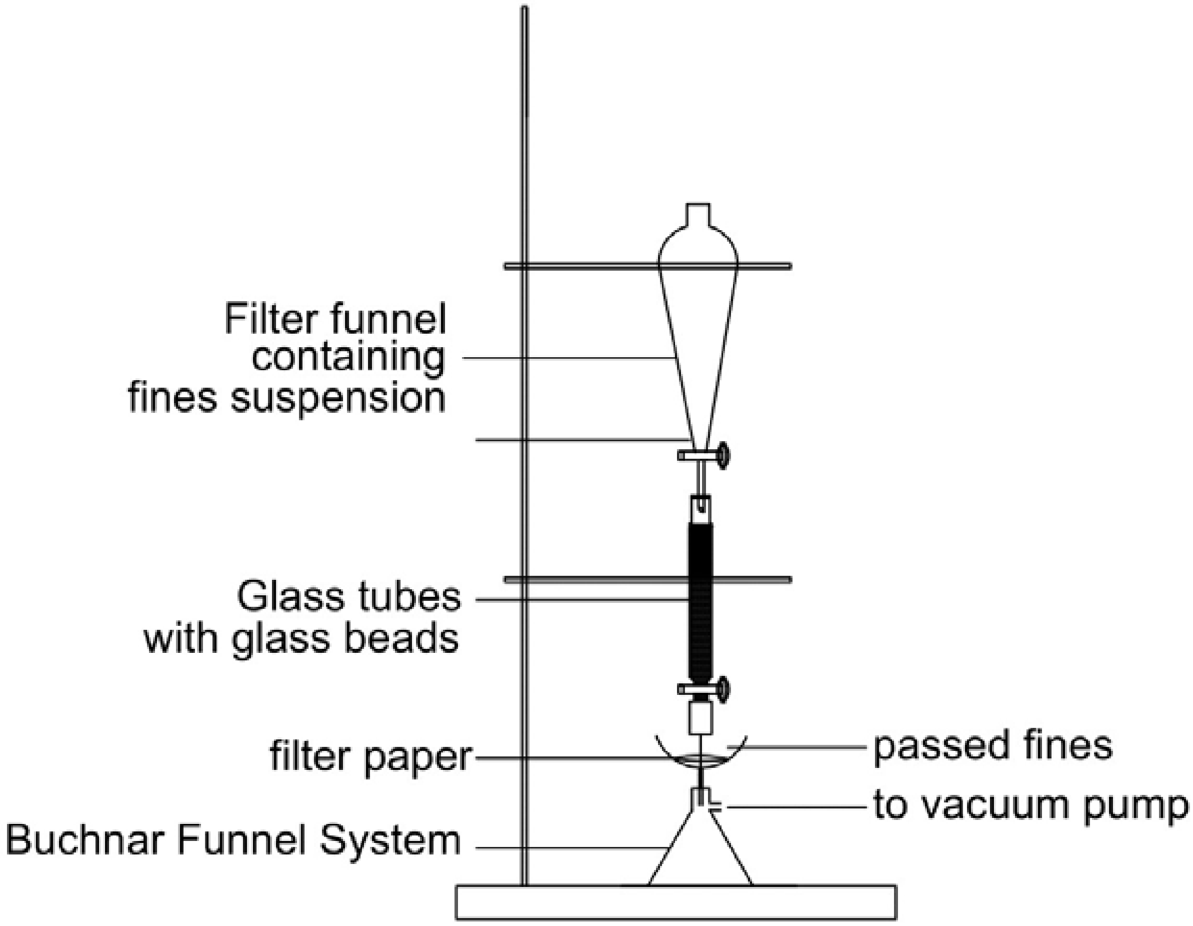


Figure 1 Apparatus set-up of fines attachment test

One of the more effective methods proposed was the application of Nano-Fluids to control fines migration. It is purposed to stop the problem before it even begins, so it serves to modify the surface properties of the pores and initiate more attraction between fines and pore walls. It works against the repulsive hydrodynamic force to maximize the critical velocity and injection rate of fluid to control the migration of fines [3]. The application of Nano-Fluids with a mass fraction of 0.1% has proven to reduce fines migration of 80% in their study. The various Nano-Fluids did not alter the zeta potential measured at the pore walls due to its negative charges; this is because of its hydrophilic nature.

OBJECTIVES

The objectives of this study are first, quantify surface forces of fines and Nano-Fluids (NF): Magnesium Oxide (MgO) and Silica Dioxide (SiO_2) through zeta potential. This study also aims to determine the best concentration of both Nano-Fluids (NF) and compare them based on their fines attachment effectiveness. It will then proceed with the identification of optimal soaking time of Nano-Fluids in sandstones to achieve a sufficient percentage (%) of fines reduction. Lastly, it formulates a more colloidally stable Nano-Fluids by adding surfactants: Triton X-100 and Tween 80.

SCOPE OF THE STUDY

This study aims to assess the compatibility and suitability of Magnesium Oxide (MgO) and Silica Dioxide (SiO_2) Nano-Fluids within Malaysian waters. The fines selected in this experiment are clays, namely illite-smectite illite, bentonite and kaolinite. They are known to be the major clay minerals in Malaysian waters. Clay minerals such as kaolinite are known to be most damaging due to its platy morphology [6]. This experimental based study highlights and investigates the solution to reduce fines migration: achieving the suitable composition of Nano-Fluids and justifying it with surface forces. Synthetic porous media is utilized with glass beads of 750-micron, to focus on the fundamental studies. These glass beads are treated by the Nano-Fluids tested and later quantified by zeta potential, a representation of the surface forces between nanoparticles and fines particles. The driving forces of the attachment are limited to gravity-driven only. These experiments are carried out in lab conditions of 25°C and 14.7 psia.

MATERIAL & METHODS

Preparation of Fines & Nano-Fluids

Before introducing fines solution to be fixated by the Nano-Fluids, an optimum migrating-condition fines concentration should be determined before the fines attachment experiment. The stability of Nano-Fluids and fines solution are different. For fines, the dry weight of the glass beaker (*g*) is measured. A 0.20% concentration of Kaolinite mineral powder is placed inside the glass beaker. The powder is suspended in 100 mL of de-ionized water (DIW), and the fines solution is placed on the magnetic stirrer at 200 rpm for 5- 10 minutes. The glass beaker is placed under the metal probe at the ultrasonic equipment that sonicates the solution for 20 minutes at 40% amplitude. Note that, this could be substituted with a bath sonicator upon availability. The glass beaker is removed and placed on a level surface and observed for 20 minutes. Only the suspended portion of the fines particles is extracted with a syringe and is used in the succeeding experiment. 10 mL is set aside into a test tube to be measured for zeta potential. The settled fines are dried completely in the oven until no moisture to measure the number of suspended fines. As for nanoparticles, the technique of preparation is similar to the fines solution.

Gravity-driven Fines Attachment Test

Fines particles used for this experiment (Kaolinite, Illite, Bentonite and Illite-Smectite) is prepared by the procedures listed in Section A. The most stable fines out of four different concentrations (0.20%, 0.50%, 1.50% and 3%) is brought forward based on their zeta potential reading. The apparatus set up, as shown in Figure 1. As a control experiment, 0.5 g of untreated, raw glass beads are weighed and washed with de-ionized water (DIW) three times and placed into the glass column. DIW is used to fill the glass column, allowing collected clear effluent at the bottom. The glass column filled with glass beads and DIW is left for 24 hours. After 24 hours, DIW is drained from the glass column. 50 mL of the selected concentration from Kaolinite, Illite, Bentonite and Illite-Smectite fines solution passes through the glass column. The stopper of the Buchner Funnel is slowly open to let the fines solution pass through the untreated raw glass beads. The effluents is filtered by using the vacuum filter (1.2-micron filtration). The fines attachment percentage (%) is then calculated based on the concentration sued earlier.

RESULTS AND DISCUSSION

Zeta Potential & Colloidal Stability of fines and Nano-Fluids

In achieving the quantitative measurement of colloidal characteristics of both fines solution and Nano-Fluids concentration, these solutions are made according to requirements. Note that the stability of both fines and Nano- Fluids represent different meanings. Fines solution needs to be stable in order to simulate migration in this research; the most stable fines solution requires the most attention from this research's area of interest. However, stable Nano-Fluids promotes better coverage of the problematic regions, where it can further travel into the production pathways.

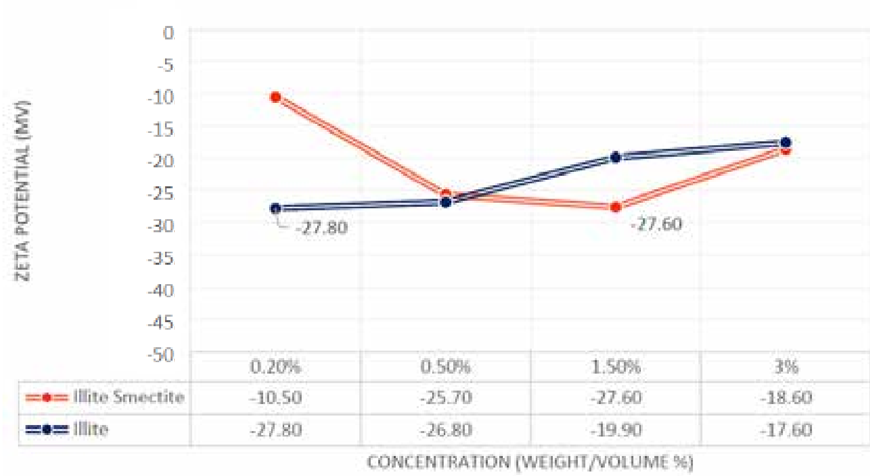


Figure 2 Graphical representation of illite smectite and illite zeta potential measurements

Figure 3 Graphical representation of bentonite and kaolinite zeta potential measurements

Comparing findings from Figure 2 and Figure 3, we can conclude that bentonite and kaolinite fines are prone to migration even at 0.35% concentration. At 0.35% concentration of kaolinite, it exhibits a zeta potential of -41.50 mV. This finding agrees with a previous study on the effects of kaolinite in fines migration, where it gives a similar value of -38 mV [18]. However, it is mentioned that at certain levels of stable Kaolinite concentrations, it might reduce the permeability more (especially when salinity is lower, with a higher pH). It concludes that kaolinite, due to its slightly spherical shape, which yields a reasonable drag force. It creates more layers of fines by migrating and deposition, leaving smaller pore size for the flow of produced fluids. Comparing illite and kaolinite fines at 0.20% concentration, it is found that illite would be more stable than kaolinite, even when kaolinite is known to be more abundant in a sandstone composition. This exciting discovery could be due to the illicit clays to move more easily and its preference to migrate according to the fluid medium [14]. The relationship of zeta potential within the selected clay minerals can be described as non-monotonic.

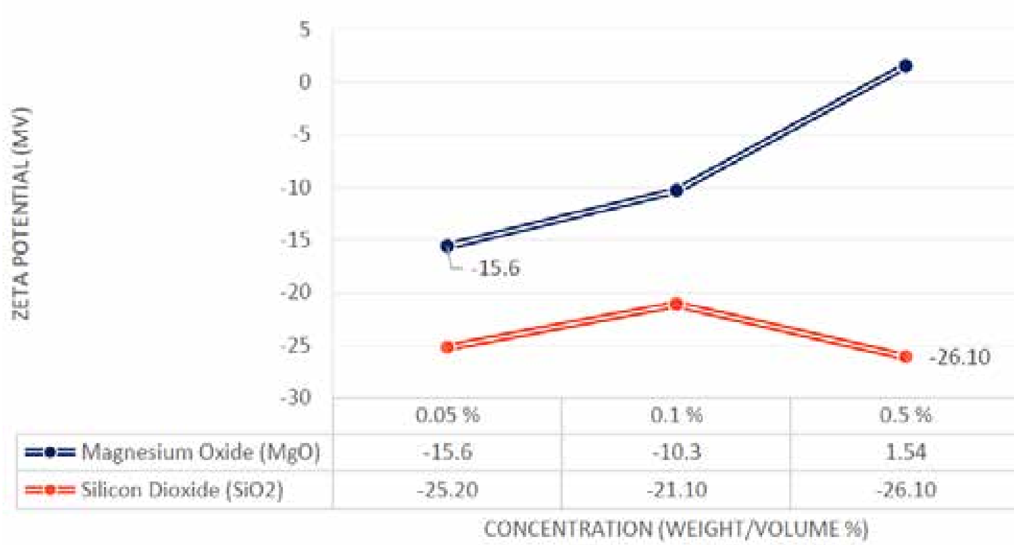


Figure 4 Zeta potential measurement of both Nano-Fluids

The lowest concentration of Magnesium Oxide (MgO) Nano-Fluid has the highest stability, as shown in Figure 4. This is supported by the concept of more commonly the increase of concentration of nanoparticle would reduce the zeta potential, making sure that more fines are attached to the given NP [19]. However, Silicon Dioxide (SiO_2) Nano-fluid might be an isolated case that differs from that pinion.

The concentration of Nano-Fluids and implications on fines reduction

The control experiments executed for all the 16 concentrations of all four fines solution enables calculation of the percentage of the attached fine from the effluents collected and dried. For the control experiment to be successful, the percentage of fines retained must be at least more than 45% to ensure that the actual experiment has significant fines to be reduced. To validate the attached fines from the untreated glass beads, we have cross-checked from the previous zeta potential measurements to ensure each fine were in the range of moderately stable. Hence, illite Smectite (1.5%) concentration, Illite (0.2%) concentration, bentonite and kaolinite of (0.35%) concentration have been selected and brought forward to this gravity-driven fines attachment experiment. From both Table 1 and Table 2, it can be concluded that the 0.1 % concentration of Nano-Fluid for both MgO and SiO_2 is the most effective in attaching fines. The values tabulated are the fines that are attached to the glass beads after that specific experiment. The control experiment with raw glass beads was run before the experiment with Nano-Fluid treated glass beads. To capture the trapping mechanism of these two Nano-Fluids, this percentage does not include the fines that continue to migrate. The illite clay fines are attached extremely well with a fines reduction of 41.49% in SiO_2 treated glass beads. It can also be inferred that the effectiveness of Nano-Fluid does not depend on the increase in concentration. Because more nanoparticles might promote agglomeration, and thus we might be introducing fines into the solution rather than aiding the reduction of the fine in the reservoir

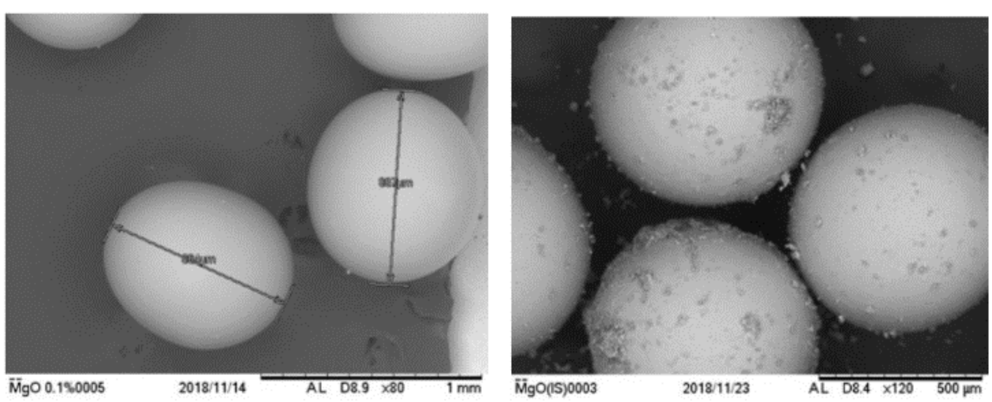
SEM imaging of a few listed conditions was initiated to take a closer look at the adsorption of the fines on nanoparticles to support the fines attachment percentage in both tables.

Table 1 Fines attached with MgO Nano-Fluid treatment

			% Finews Reduced by MgO		
Fines	**Concentration**	**Control Exp.**	**0.05%**	**0.1%**	**0.5%**
Illite Smectite	1.5%	65.41%	87.46%	61.65%	58.32%
Illite	0.2%	57.31%	96.80%	97.60%	94.01%
Bentonie	0.35%	51.88%	70.41%	79.71%	61.005%
Kaolinite	0.35%	72.82%	77.54%	97.75%	93.26%

Table 2 Fines attached with SiO_2 Nano-Fluid treatment

			% Finews Reduced by SiO_2		
Fines	**Concentration**	**Control Exp.**	**0.05%**	**0.1%**	**0.5%**
Illite Smectite	1.5%	65.41%	61.65%	70.13%	67.92%
Illite	0.2%	57.31%	98.80%	98.80%	96.41%
Bentonie	0.35%	51.88%	48.86%	62.81%	62.55%
Kaolinite	0.35%	72.82%	66.32%	97.75%	84.98%



a) b)

Figure 5 (a) MgO NP treated glass beads, (b) MgO NP treated glass beads with Illite fines attached

Due to the performance of the fines attachment percentage (%), this study has evaluated a 0.1% concentration of both Magnesium Oxide and Silicon Dioxide. This research will first observe from the attachment coverage of fines before moving closer to investigate what was attached. Comparing Figure 5a and Figure 5b, it can be observed visually that the Illite fines have been distributed evenly around the NP treated glass beads with before and after effect. Even during the fines attachment test, the glass beads recompressed to simulate sandstone grains being packed. Figure 5b shows that the attachment of these fines in between the synthetic grains has a certain distance that allowed the flow of fluids over the trapped and fixated fines. These nanoparticles used, namely MgO and SiO2, have unique abilities to be chemically stable and suitable interaction potentials. Due to their small sizes, they can cover a larger area of interest, which is shown by how fines are trapped on the surface of the glass beads easily.

From Figure 6a, the combination of "cornflake" or "oakleaf" structure of smectite clay can be seen on the bottom right, while the illite clays that have curved, long edges and platy looking can be seen on the left portion SEM image [16]. The Kaolinite fines attached to the Silicon Dioxide treated glass beads have appeared to clump and accumulate at a specific area, while other certain areas might have a slightly thin layer of fines attachment, as seen in Figure 6c. Figure 6d, the fines is zoomed in to investigate further due to the distance of interactions of NP and fines, as illustrated in the DLVO theory. As supported and agreed by a previous study, it was approximately 0.5 nm the Born repulsion energy will slightly dominate more attractive forces, and the attraction begins around 1 nm [18]. However, this study used a concentration of 0.05% for their nanoparticles and varied injection rate to prove the negligence of hydrodynamic potential due to the fact or large particles and higher injection velocities that is beyond the scope of this research.

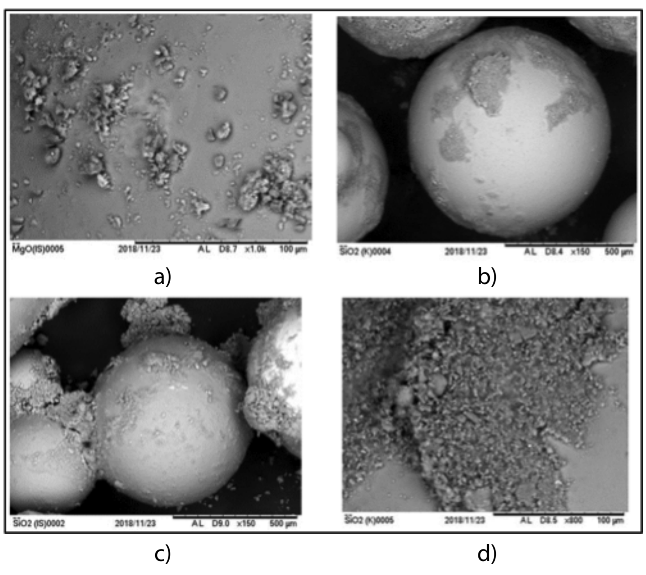


Figure 6 a) closer visualization of attached Illite Smectite fines, b) Fines attached on SiO_2 NP treated glass beads, c) Kaolinite fines attached in SiO_2 NP treated glass beads, d) Visualization of Kaolinite Fines

The pseudohexagonal crystals of kaolinite can be fixated on the Silica Dioxide treated glass beads. Note that these synthetic grains have been dried for SEM imaging. The amount and distribution of attached fines might appear to be slightly different under immersed conditions.

Variation of soaking time

This objective is initiated to account for the field application of NP treated sandstones to trap the migrating fines. To provide a time-efficient solution, the optimal time for both Nano-Fluids to adhere to the surface of the glass beads must be determined.

As proposal by a similar study, the coating of NP on the sandstones of problematic zones can be conveyed through the hydraulic fracturing in the proppants. It could also be injected separately to only coat the drainage zones [18].

From the conducted experiment of a constant fines solution (Illite) variated between both types of Nano-Fluids, Magnesium Oxide (MgO) seems to achieve optimum performance at two hours of soaking while Silicon Dioxide (SiO_2) takes one hour. It must be taken into consideration that this is a unique case for a specific clay mineral, which might differ from other cases.

The significance of this objective is to reduce the time used to prepare and execute the treatment job for the candidate reservoir because for every hour, the well is shut-in for treatment. It would be considered as non-productive time (NPT). The setting time for nanoparticles into the sandstone formation need to be minimized, so the results and effectiveness of how it reduces fines migration can be observed. The results of the soaking time and efficiency as tabulated in Table 3. The percentage of reduced fines by comparing the controlled and real experiment is shown in Table 4.

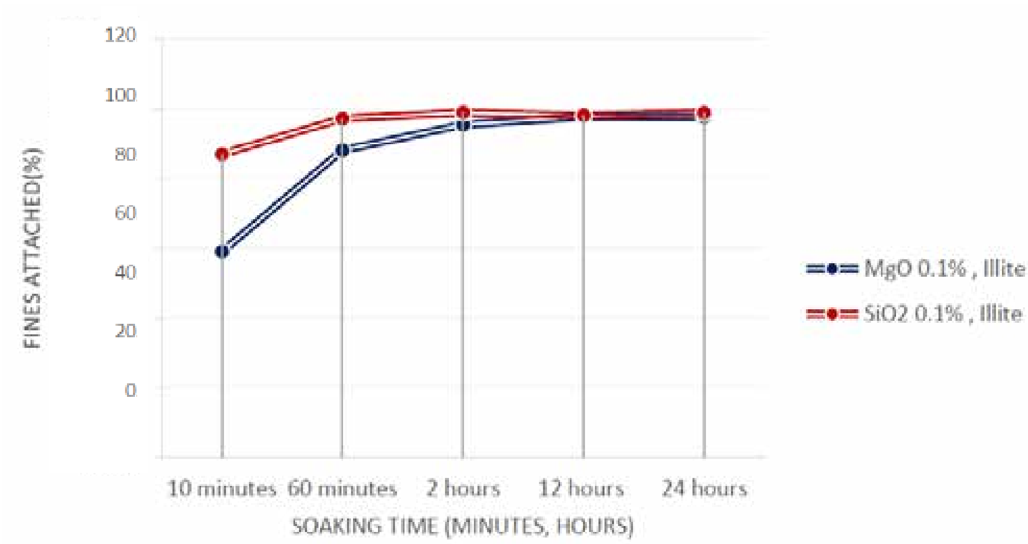


Figure 7 Effect of soaking time on the fines attachment efficiency

Table 3 NP Soaking time variation and attachment percentage

No.	Time (minutes, hours)	NF and fines solution	
		MgO 0.1%, Illite	SiO_2 0.1%, Illite
1	10 minutes	59.00%	86.92%
2	60 minutes	88.00%	89.53%
3	2 hours	95.34%	98.85%
4	12 hours	97.63%	98.00%
5	24 hours	97.60%	98.80%

Table 4 Fines reduction percentage

No.	Time (minutes, hours)	NF and fines solution	
		MgO 0.1%, Illite	SiO_2 0.1%, Illite
1	10 minutes	1.69%	29.61%
2	60 minutes	30.69%	32.22%
3	2 hours	38.03%	42.54%
4	12 hours	40.32%	40.69%
5	24 hours	40.29%	41.49%

Effect of surfactant TX-100 and Tween 80 on NP stability

Fixing the concentration of Nano-Fluids at 0.1%, this research has proceeded to test if the addition of surfactant might be able to aid in increasing the stability of the Nano-Fluid. The process of which is executed is critical because, without the proper dispersion of the nanoparticles itself, the surfactant might be surrounding an agglomerated portion of the nanoparticle powder suspended in de-ionized water.

With the reference of -15.6 mV for Magnesium Oxide (MgO) and -26.10 mV for Silicon Dioxide (SiO_2) for this experiment, only Silicon Dioxide manage to achieve the objective of increasing its stability. It has managed to attain -29.43 mV with five drops of Tween 80 and -30.6 mV with five drops of Triton X-100. It can be concluded that TX-100 and Tween 80 might work better with Silicon Dioxide than it is compatible with Magnesium Oxide (MgO). Enormous zeta potential was found when mixing extremely diluted surfactants with the Nano-Fluids; these values are outside the range of standard classifications. Therefore, it was suspected that the surfactants were not able to coat any nanoparticle, as the solution was also visible. Since these were outliers, we will not graphically present these measurements. From Figure 8, the positive reading of Magnesium Oxide (MgO) followed by an almost similar magnitude negative reading of zeta potential, shows that the zeta potential has crossed the isoelectric point (IEP). Due to the weak performance of Magnesium Oxide (MgO) with both TX-100 and Tween 80, it is not brought further into the mixing of 10 drops of surfactants, as depicted in both graphical figures. This could also mean that an abundance of surfactant- nanoparticle ratio might lead to a flipside outcome due to the drag forces that might be more significant when the nanoparticle is being coated with a surfactant.

From Figure 9, Tween 80 might be slightly more compatible with both Nano-Fluids but best with Silicon Dioxide, as mentioned before. However, the decrease in zeta potential (stability) with an increased concentration of Tween 80 might imply that the coating of surfactant has weighed down the silica nanoparticles. It was previously studied that approximately 14 micelles of surfactant are adsorbed on a single silica particle. Concerning the short particle distance between surfactant- nanoparticles, a short-range inter-particle repulsion might make the aggregation reversible and potentially leads to another kind of interaction.

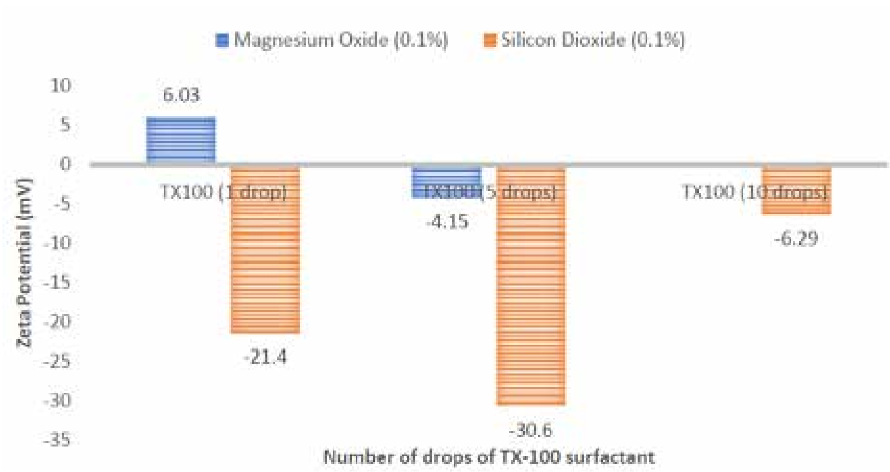


Figure 8 Zeta potential of Nano-Fluid with the addition of Triton X-100

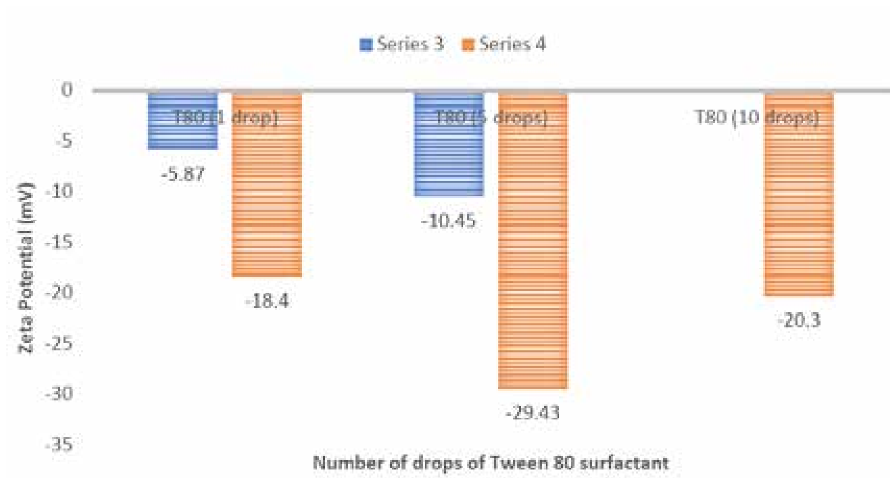


Figure 9 Zeta potential of Nano-Fluid with the addition of Tween 80

CONCLUSIONS

From this research, we can draw certain conclusions that may be helpful to others who are interested in nanotechnology and its role in fines migration reduction. The evaluation of the surface forces acting between Nano-Fluids and fines clay minerals of illite-smectite, Illite, bentonite and Kaolinite has been carried out. Bentonite and Kaolinite fines are more susceptible to migration in sandstone reservoirs due to their high colloidal stability from their zeta potential measurements and their morphologies. The unique characteristics of bentonite being one of the most prominent montmorillonites and Kaolinite mineral's dominance in most sandstones have both played roles in supporting their higher possibilities of migration. Hence, more attention should be given when the composition of the fine is abundant in these minerals. It can also be concluded that Silicon Dioxide (SiO_2) is more stable than when these nanoparticles are suspended in de-ionized water only. It gives zeta potential values of -26.10 mV, which falls into the moderately stable classification.

The gravity-driven fines attachment test has clearly shown that the performance of 0.1% weight/volume the concentration of Nano-Fluid outperforms the other two concentrations. The reduction of fines was as much as 40% overall; however, this was ensured by making sure that the attached fines from the untreated, raw glass beads were sufficient to quantify. This was then further evaluated and observed through SEM imaging that confirms the pattern of which the fines attached themselves unto the synthetic sandstone made of glass beads. To enhance this application for a field aspect that prioritizes time and efficiency, we varied the soaking time of Nano-Fluid in sandstones. We have found that Magnesium Oxide (MgO) takes Approximately two hours to achieve optimum performance, while Silicon Dioxide (SiO_2) is slightly faster with an hour soaking time. The addition of surfactants (Triton X-100 and Tween 80) has also shown significance in this study. Through the evaluation of zeta potential once again, Tween 80 has shown to be more compatible with the selected Nano-Fluids than Triton X-100. It increased Silicon Dioxide (SiO_2) Nano-fluid zeta potential value to be highly stable at -30.6 mV, which is beneficial for the Nano-Fluid to travel deeper into the formation to treat problematic areas of fines migration. The research design has been tailored to the suitability of our experimental materials and availability while weighing the cost and time to achieve our objectives. For instance, Particle Size Distribution (PSD) was chosen over Photon Correlation Spectroscopy (PCS) in the determination and characterization of nanoparticle size because PCS requires a very dilute suspension sample and very sensitive to impurities – which in this case, effluents of our samples are highly un-pure with no definite viscosity.

RECOMMENDATIONS

This study recommends having a variety of different clays and other fines materials. All these done by having field data of fines composition of a specific location. Smaller intervals of Nano-fluid concentrations should be investigated to achieve an optimum level of fines attachment. Next, more studies should be carried out to attempt more stabilization techniques of Nano-Fluids, e.g., using another dispersant to functionalize, variations pH of the solution. Further investigation should be made to understand the number of surfactants to be used in stabilizing the Nano-Fluids.

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