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$Gr\text{-}Al_2O_3$ Nanoparticles based Multi-Functional Drilling Fluid

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Abstract Various chemical additives are used in drilling fluids to improve the drilling efficiency and increase oil production. Dispersing nanoparticles as additives in drilling mud is one of the promising techniques that could help solve many difficulties in drilling operations. However existing work is insufficient with numerous inconsistent outcomes. This study aims to discover and evaluate the impact of graphite-alumina hybrid nanoparticles on the rheology, thermal properties, zeta potential and electrical conductivity of water-based drilling fluids. It shows that that the addition of nanoparticles improved the effectiveness of water-based mud (WBM) both at static and dynamic situations due to the establishment of an appropriate gel structure that can be easily broken under low shear stress. Furthermore, these nanoparticles improve the degree and the speed of the structural recovery by reducing the relaxation time, which is necessary to avoid the sedimentation of weighting materials and drilled cuttings. Besides, nanoparticle seeded fluid improves the effective thermal conductivity to conventional drilling fluids, and the highly charged suspension resulting from nanoparticles addition is desirable for the drilling operation. It is also observed that nanoparticles could penetrate to the porous wall of the wellbore and clog the pores, forming a thin and impermeable cake that reduces drilling fluid loss. Properly engineered, nanoparticles could improve the performance of conventional drilling fluids significantly.

Keywords: Drilling fluids; nanoparticles; rheology, thermal conductivity, multi-functional

1. Introduction

The global demand for energy is expected to increase as much as 50% in the next 20 years, and the demand for oil and gas will also increase[1]. The area of finding "easy oil" is coming to an end, and future supply will become more reliant on hydrocarbons produced from unconventional hydrocarbon sources and enhanced oil recovery (EOR) processes. The performance of a drilling fluid is essential for the smooth and stable production of hydrocarbons. Drilling fluid is an essential element in the well-construction process that remains in direct contact with the wellbore throughout the entire drilling operation. A well-constructed and reliable wellbore can significantly reduce the nonproductive time. The design of a drilling-fluid system is central to achieve this objective[2]. Drilling fluids should be formulated to work appropriately under expected wellbore conditions, possessing good rheological and thermos-physical properties. Quite recently, considerable attention has been paid to using nanoparticles to improve drilling fluid performance[3, 4, 5]. A considerable effort has been recently devoted to employing nanoparticles in drilling fluids to control the mud filtrate volume[6, 7, 8, 9], minimizing differential pipe sticking[10], improving drilling and production at high pressure and high temperature (HPHT) conditions[5, 11, 12, 13], enhancing shale stability[4, 14] and improving rheological properties[8, 9, 15]. Nanoparticles were used by some researchers to develop thermal, electrical and HPHT rheology of water-based mud[12, 16]. It is detected that the increase of nanoparticle concentration promotes thermal and electrical features of drilling fluids. The results of recent studies showed that nanoparticles enhanced thermal and electrical characteristics by approximately 35% compared to water-based mud (WBM). These studies have shown that the addition of nanoparticles could improve some properties of the drilling fluid, which shows some promise for future applications. On the other hand, a few researchers have shown the impact of nano-composites for drilling mud application[17, 18]. Nanoparticles were used as fillers for polymer composites due to their high thermal and electrical conductivities[17, 19]. It is expected that the addition of small amount of nanoparticles could actively improve the thermal, electrical and mechanical properties of a polymer matrix as used in a drilling fluid[17, 19, 20].

However current state-of-art studies have a number of limitations, as commented here. Firstly, the understanding of the properties of nanoparticle-based drilling fluids is insufficient. Most of the studies have been focused on one property, such as rheological property or filtration property[13, 21]. As a drilling fluid provides multi-functions during the drilling operation, such as cooling, lubrication and cutting transport. Addition of nanoparticles would clearly affect these functions, and it is essential to understand the variations of related properties having a complete assessment of the nanoparticle effects. Secondly, the impact of nanoparticles is multi-facets, and their influence on different properties are different. The addition of one nanoparticle may improve one property but may not improve the other[22]. It is unlikely that the addition of one type of particle could improve all related properties. The search for right nanoparticle or hybrid particles is still ongoing. Thirdly, the drilling fluid cycle is often unsteady; conjointly variate between a movement and a steady state. Therefore, the challenge is to design a single recipe of drilling fluid that exhibits agreeable abilities both at static and in dynamic conditions. However, most of the previous studies do not take that into account. Finally, the drilling fluids behave as viscoelastic materials, exhibiting both viscous and elastic behavior , but the study of the viscoelasticity behavior of Nano-drilling fluid is insufficient in the literature[23]. Identifying the differences in viscoelastic response could lead to better design of a drilling fluid recipe. Aiming to address these limitations, this work develops a novel hybrid nanoparticle system, graphite-alumina (Gr-Al_2O_3), dispersed in a WBM, and examines their properties under both dynamic and static conditions. The Gr-Al_2O_3 nanoparticles can exhibit a notable enhancement in drilling fluid physicochemical, mechanical and thermal properties due to large surface area/volume ratio that offered by these nanoparticles and their high thermal and electrical conductivity. The rheological, electrical and thermal properties of the newly formed drilling fluids are examined to reveal their potential functions. Al_2O_3 nanoparticles are selected due to their ability to enhance rheological, thermal and electrical properties of drilling muds. Graphite nanoparticles are chosen to provide less filtrate loss due to there morphology. In addition, Gr-Al_2O_3 mixture is considered as non-hazardous and suitable for use in environmentally sensitive locations and applications

2. Experimental Work

2.1 Preparation of Nano Drilling Fluids

The drilling fluids were prepared in the laboratory according to a general mud composition. A typical formulation includes 335ml of water and 20g of sodium bentonite particles. The sodium bentonite was purchased from Mistral Industrial Chemicals Company. The physical and chemical characteristics of sodium bentonite are shown in Table 1. They were mixed by overhead high shear speed mixer for 20 minutes until complete hydration of the bentonite.

Table 1. Physical and chemical characteristics of sodium bentonite

Typical Chemical Analysis		Typical Mineralogy		Other Typical Properties	
SiO_2	57.1%	Montmorillonite	92%	Bulk Density	800 – 900 Kg/m^3
Na_2O_3	3.27%	Calcite	4%	Swelling Volume	29 ml/2g
Al_2O_3	17.79%	Feldspars	2%	Moisture	Maximum 14%
K_2O	0.9%	Quartz	1%	Cation Exchange Capacity	78 meq / 100g
Fe_2O_3	4.64%	Dolomite	1%	Sieve Analysis	Maximum 5% retained on a 150µm sieve
TiO_2	0.77%				
CaO	3.98%				
Mn_2O_3	0.06%				
MgO	3.68%				
LoI	7.85%				

A mixture of alumina (50%) and graphite (50%) nanoparticles at different concentrations (0.0, 0.2, 0.4, 0.6, and 0.8 wt %) were dispersed, mixed by a Hamilton beach mixer for 20 minutes into the formed mud. These nanoparticles are obtained from NanoTek® and Nanoshel respectively. Their morphology was identified by a scanning electron microscopy (Hitachi SU8230: high-performance cold field emission (CFE) SEM), as shown in Figure 1. The SEM on the left illustrates graphite particles exhibits a layered and needle structure morphology. The SEM on the right demonstrates the rod-like morphology of Al_2O_3 nanoparticles. The Morphology of the particles was also characterized by a transmission Electron microscope (FEI Tecnai TF20 TEM) and is shown in Figure 2. The TEM shows the same particles morphology that indicated by using SEM.

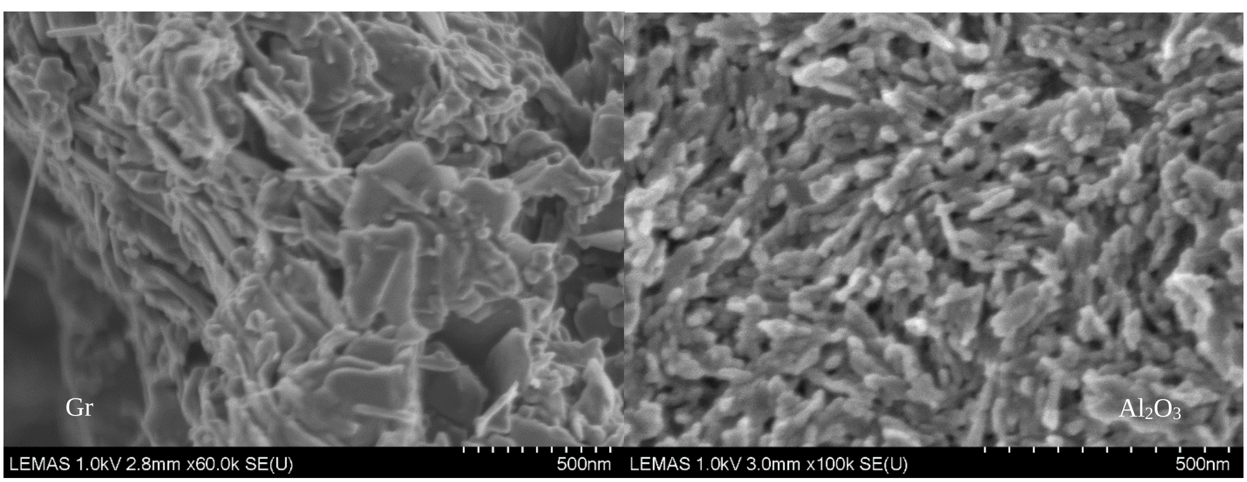

Figure 1. Scanning electron micrographs of graphite and alumina.

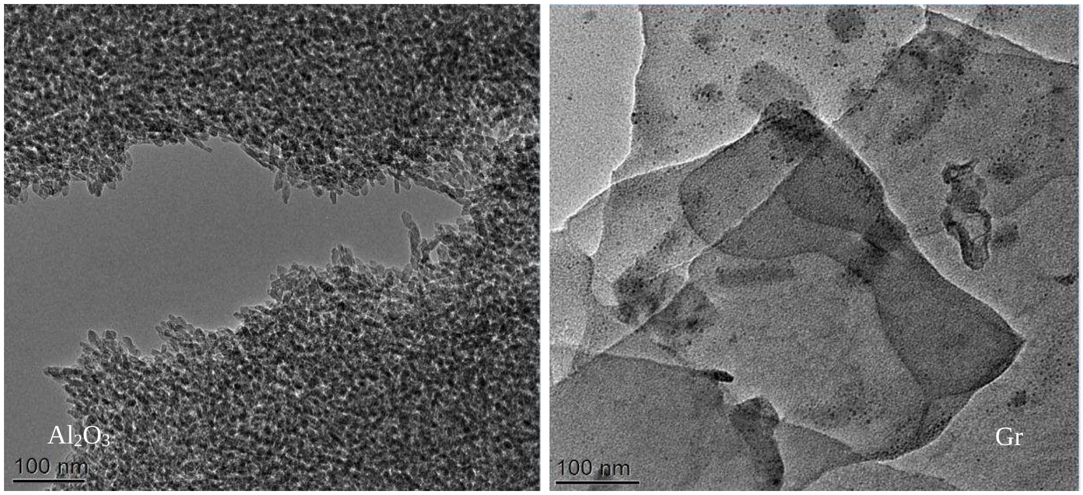

Figure 2. Transmission Electron Microscope (TEM) images of graphite nanoparticles and aluminum oxide nanoparticles.

The hydrodynamic nanoparticles size distribution in distilled water was checked by using a Malvern Nanosizer based on the dynamic light scatting (DLS) method (DLS, Mavern Zetasizer, Marvern, UK), which displays an average size of 198nm, peaking at 221nm for Al_2O_3 and average size 309nm with two peaks at 80nm and 400nm for Gr (Figure 3).

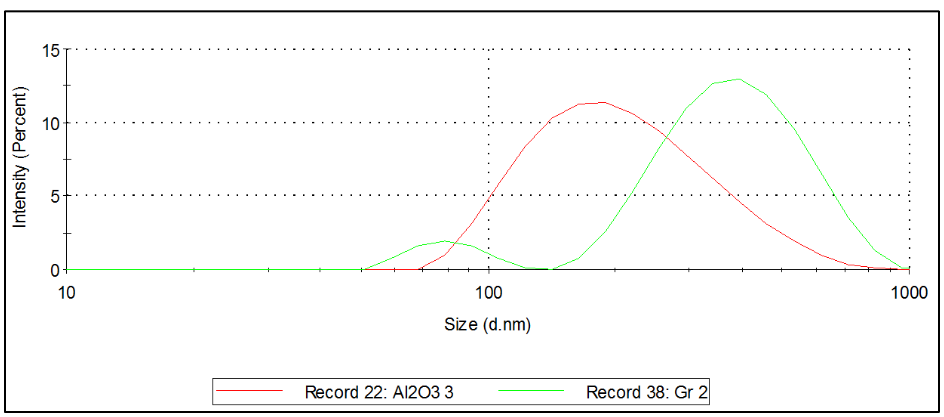

Figure 3. the particle size distribution of graphite and alumina in distilled water measured by the DLS method.

2.2 Experimental apparatus

In general, Fann 35 Viscometer is generally used to measure the rheology of the drilling fluid in most oil fields across the world[24]. However, in this research, rheology measurements are done by the Anton Paar MCR301Rheometer at ambient conditions

(Figure 4, left side). This Rheometer is more precise and more advanced: it can measure viscosity and related attributes. The MCR301 was used to do more analysis of fluids while Fann 35 failed[25]. The flow behavior of drilling fluids was investigated by using a rotational system. The oscillation amplitude sweep test was employed to find the linear portion of the viscoelasticity and to observe the structural characteristics of fluids, While the combination oscillation-rotation-oscillation was utilised to study the thixotropy. The measuring system consists of a rotating cylinder called bob and cylindrical cup, where the fluid rests. In a working situation, the cylindrical bob is rotated inside the cylindrical cup which is filled with fluid. The mud volume required for the test is determined by a marker inside the cylindrical cup.

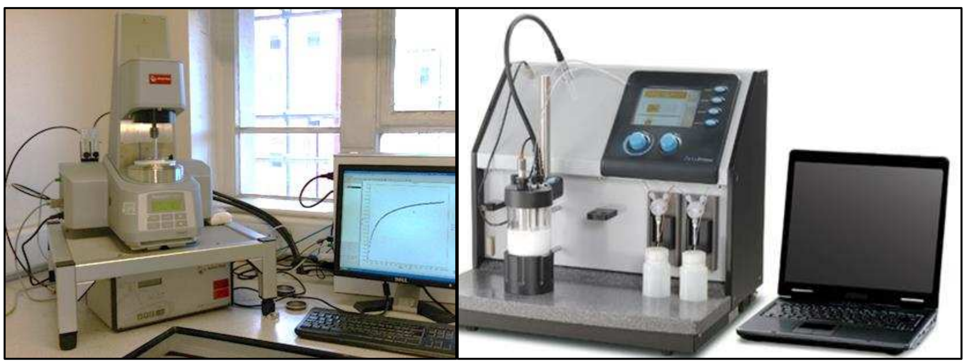

Figure 4. Anton Paar MCR301 Rheometer (left side) and Zeta probe (right side).

The KD2 Pro Thermal Properties Analyzer made by Decagon Devices, Inc. has been employed to conduct the thermal conductivity of the prepared Nano-drilling fluids with the accuracy of 5%. The KD2 Pro is a handheld device, battery-operated, menu-driven device which works based on the transient heated needle to conduct thermal properties of solid and fluid media[26]. The thermal conductivity measurements were done by totally oriented the KD2 pro KS-1 a sensor with 60 mm in length and 1.3 mm in diameter vertically in 40ml of drilling fluid sample. The measurement was done on a vibration isolation table at overnight after turning off any source that cause vibrations to prevent the errors from convection.

The Zetaprobe manufactured by Colloidal Dynamics, LLC has been used to measure the zeta potential of various Nano-drilling fluids without dilution. The Colloidal Dynamics

Zetaprobe worked based on the electroacoustic method which offers straight measurement (without dilution) for colloidal dispersions with moderate and high particle concentrations while the other techniques necessitate sample dilution and sample preparation which are both time-consuming and error-prone. The Zetaprobe builds from a compact design with built-in titration, a versatile dip probe sensor, and software wizards as in figure (4, right side). The Zetaprobe measured the Zeta Potential, Dynamic Mobility, conductivity, temperature and pH of the stirred sample at a controlled speed. All functions are controlled via a Graphical LCD user interface, and the instrument also interfaces to and can be controlled from a PC.

3. Results and Discussion

3.1 Rheology

Rheology is a crucial parameter of drilling-fluid performance. Modification, the rheology of drilling fluid, can be a convenient solution to most drilling difficulties like pipe sticking, loss circulation and formation damage. The drilling fluid behaves as viscoelastic materials. This confirms that drilling fluids display both viscous and elastic behavior . Therefore, it’s essential to investigate the viscoelasticity behavior of drilling fluids to understand how additives can develop the structure of drilling fluids. Identifying the differences in viscoelastic response could lead to better design of a drilling fluid recipe. The speed of the structural recovery (thixotropy) is also essential. To evaluate flow, viscoelastic and thixotropic properties of drilling muds and it is regularly suitable to deal with them separately, some rheometric assessments that can be conducted on a rheometer.

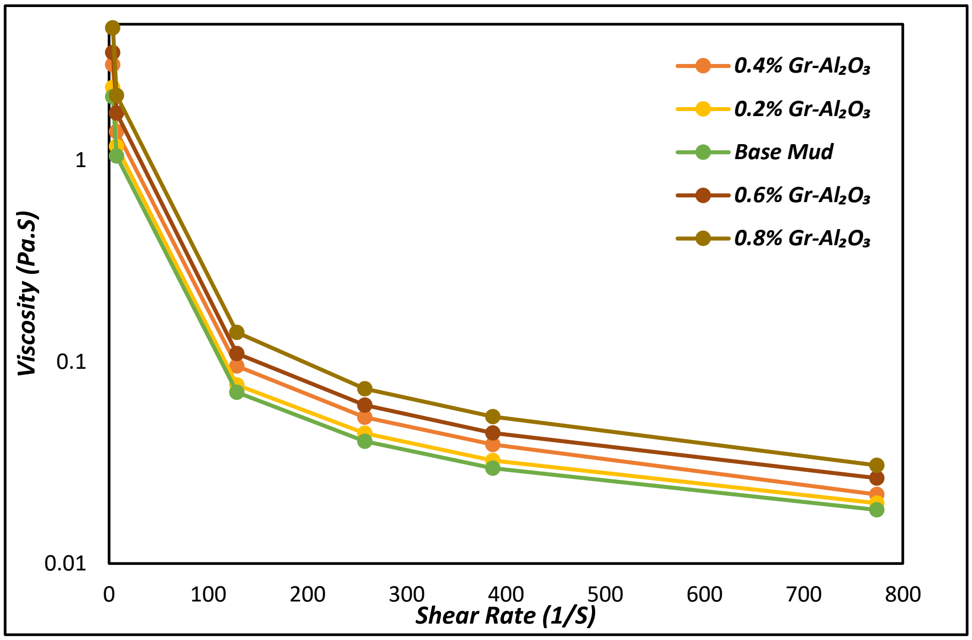

Figure 5. Viscosity of the prepared drilling fluids as a function of shear rate.

The first set of analyses investigated the flow curves, which usually use the graphical description of flow behavior . It exhibits the flow behavior at low shear rates as well as high shear rates. The viscosity of drilling fluid decreases with increasing shear rate. This flow behavior is called shear-thinning as in figure 5. Therefore, it is not sufficient to indicate only the viscosity. It is consistently necessary to designate also the shear rate that was utilised for the mensuration. It is interesting to note that the viscosity of drilling fluid is increased with hybrid nanoparticle addition. The drilling fluids viscosity is increases by ~10% when only 0.2% of Gr-Al_2O_3 is added to 350mL of the base drilling fluid. In addition, about 32% viscosity increment was measured when 0.4% of Gr-Al_2O_3 was added in the same conditions. Furthermore, around 55% viscosity enhancement was noted when 0.6% of Gr-Al_2O_3 was added, as revealed in Figure 5. The reason behind that could be due to the nanoparticles shape with large surface to volume ratio. This also could be linked with the randomly dispersion of nanoparticles on the surface of bentonite particles and award links between bentonite particles, which might promote gelation of the bentonite. These results are consistent with previous findings in the literature[6, 13, 27].

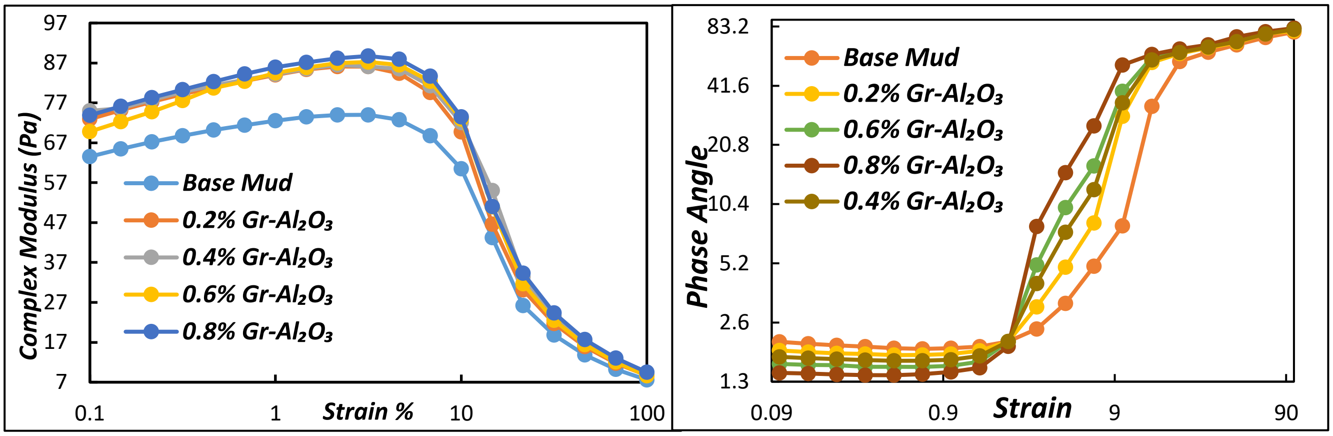

Figure 6. Complex modulus and phase angle of the prepared drilling fluids as a function of deformation percentage.

Characteristics such as loss and storage modulus, complex modulus, dynamic yield point, structural stability, and phase angle describe the viscoelastic behavior of drilling fluids. The inter-particle interaction plus the particle networks in the drilling fluids can be measured by noting the passage from a solid-like condition to a liquid-like status. Presentation of variation complex modulus (overall resistance to deformation) as a function of strain obtained through oscillatory amplitude test is used as a suitable tool for investigation of the strength of the particle connection in the fluid (the stiffness of material; the higher the modulus, the more stringent the material)[23].

As clear from Figure 6, the complex modulus increases with increasing nanoparticles concentration. This means that nanoparticles give more elastic properties to the fluid before flow point and this what we need since this state helps retain cuttings and weighting materials suspended in the drilling fluid instead of allowing for settling into the wellbore. The flow point and the degree of viscoelasticity can be much more easily be presented by plotting phase angle δ against shear stress. When an oscillatory (sinusoidal) deformation (stress or strain) is applied to a sample. The material response (strain or stress) is measured. The phase angle δ is the phase shift between the deformation and response. The value of yield stress (flow point) can be accounted by interpolating the stress at which the phase angle is 45° (G" = G'). A low value of the phase angle δ represents a greater value of G' compared to G". This means that the drilling fluid with lower phase angle is better develop than with higher phase angle. As indicated in the figure 6, the addition of Gr-Al_2O_3 nanoparticles conferred drilling fluids more elastic properties at linear viscoelastic range (LVE) and less shear stress at flow point. This means that Gr-Al_2O_3 drilling fluids have better gel structure and this gel can break with less shear stress as shown Figure 6. The results also indicate that nanoparticles can develop the structure of drilling fluid. Moreover, even nanoparticles give the more elastic property to drilling fluid; the yield stress is still same. This means that Nano drilling fluids display sufficient abilities both at rest and in the movement since they have better gel structure and this gel can break with same shear stress. When drilling fluid pumps are shut off or running at very low speed, the drilling fluid will take on a gel-like state. This state of the drilling fluid helps retain cuttings and weighting materials suspended in the drilling fluid instead of allowing for settling into the wellbore. It is appropriate that the drilling fluid quickly develops high gel strength to resist the settling of massive particles out of suspension. These results This concurs well with [28] and also confirms our previous findings[22].

The laboratory results also proved that the addition of hybrid nanoparticles could develop the thixotropic properties of drilling fluids. As clear from Figure 7, the increase of nanoparticles concentration secures more elastic behavior to the drilling fluid and increase the degree and the speed of the structural recovery through reducing the time required for rebuilding. It is preferable to minimize the reconstruction time to prevent the sedimentation of the weighting materials and drilled cuttings. Our findings appear to be well consistent with previous results[29, 30].

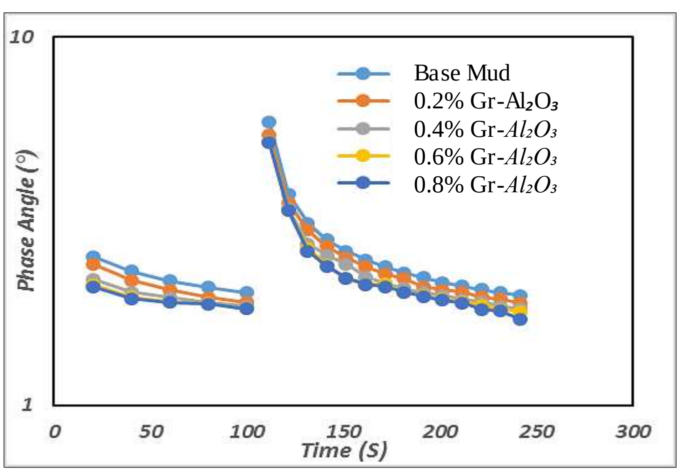

Figure 7. Thixotropy of the prepared drilling fluids as a function of time and shear rate.

3.2 Thermal conductivity

The KD2 Pro Thermal Properties Analyzer has been standardized by using distilled water before the measurements. The thermal conductivity measurements were done at the ambient temperature. The thermal conductivities of the prepared drilling fluids as a function of nanoparticle concentration are investigated. Figure 8 show the thermal conductivity improvement as a function of nanoparticles concentration for base water-based fluid at room temperature. The mass fraction of the nanoparticles is varied from 0 % to 0.8 %. It is found that there is a linear relationship of the thermal conductivity with variation in the concentration of the nanoparticles. The linear trend was observed in the literature[31] between nanoparticles concentration and thermal conductivity. Results indicate that an increase in the nanoparticle concentration enhances the thermal conductivity. The laboratory results illustrate that the water-based drilling fluid thermal conductivity is improved by around 10% in the presence of 0.8 wt% Gr-Al_2O_3 at room temperature. The increase in the thermal conductivity of the nano-drilling fluids is due to high specific surface area of the nanoparticles. The presence of nanoparticle sizes causes an increase in the surface area per unit volume of the particle. As heat transfer is a function of surface area, it ultimately increases the effectiveness of the nanoparticles to transfer the heat to the base fluid[30, 32]. An improved drilling fluid thermal conductivity is an indication of the ability of the fluid to cool faster as it moves up to the surface. On the other hand, with enhancement the heat transfer between the drill bit and drilling fluid, the bit remains cooler and this possibly helpful for innovative design of drilling muds for high temperature and high pressure (HTHP). The outcomes of effect of Gr-Al_2O_3 nanoparticles are consistent with previous findings in the literature[33].

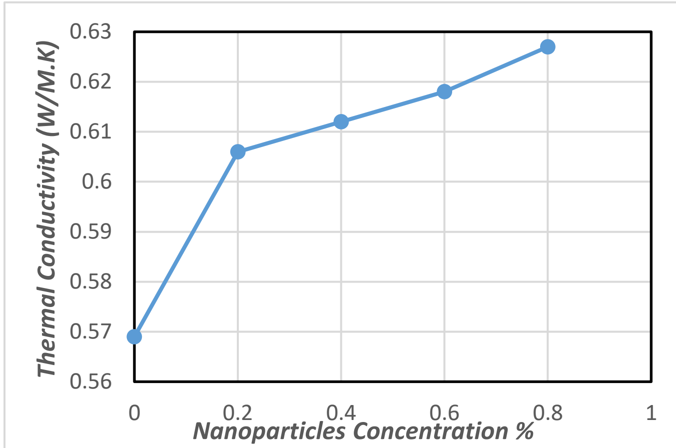

Figure 8. Thermal conductivity of prepared drilling fluids as a function of nanoparticle concentration.

3.3 Electrical conductivity

Table 2 records, the electrical conductivity values of water-based muds measured at each nanoparticle's concentration. It is noticed that the addition of Nano-particles to base mud enhances the electrical conductivity. The laboratory outcomes indicate that the water-based drilling fluid electrical conductivity is improved by around 4% in the presence of 0.2 wt% Gr-Al_2O_3. The enhancement of the electrical conductivity is observed to be from about 4 to 8.4% when the concentration of Gr-Al_2O_3 increased from 0.2 to 0.8 wt% due to increase in nanoparticle concentration cause increase interaction between nanoparticles which resulted in an enhancement in the electrical conductivity [34]. The enhancement of the electrical conductivity of fluids by nanoparticles may be attributed to the formation of electrical double layers (EDL). When nanoparticles are dispersed into the base fluid due to the EDL effect, the surface of these particles gets charged and can transfer charge to the solution. Therefore, the electrical conductivity of a nanofluid can be related to the concentration of these carriers (nanoparticles). As the nanoparticle's concentration increased the number of charge carriers and then the electrical conductivity increased. The previous findings reveal considerable enhancement of electrical conductivity property with the addition of nanoparticles to the base fluid [35] and this supported this research outcomes. The electrical conductivity has a significant influence of the nature of the drilling fluid upon electric logs (resistivity imaging) as the drilling fluid must be the medium through which the electric logging device is made to traverse the formations. Electrical logs have been used to distinguish the differences between shales, sandstones, limestones, and other rock forms readily. Besides electric logs have been found, in some instances, to be useful in estimating the nature of fluid contained within the pores of a rock. The electrical conductivity of a drilling mud, its filtrate, and filter cake are three of its more important qualities, as far as electric logging is concerned.

Table 2. Electrical conductivity measurements of drilling fluids at 20 °C (mS/cm)

Fluid Samples	Electrical Conductivity	Improvement
WBM	2.84	0
0.2 wt% Gr-Al_2O_3	2.93	3.2
0.4 wt% Gr-Al_2O_3	2.97	4.6
0.6 wt% Gr-Al_2O_3	3.01	6
0.8 wt% Gr-Al_2O_3	3.08	8.4

3.4 Zeta potential

The nanoparticles charge, and zeta potential play a significant role in the performance of the rheological behavior. The nanoparticles with a positive charge displace the dissociated cations from the surface of bentonite which result in a different clay platelet structure, yielding higher yield stress values[8, 36]. While the nanoparticles with a negative surface charge increase the repulsion forces between the clay platelets, which leads to deflocculating of the platelets and, thus, higher viscosities and weaker yield structure. Sodium montmorillonite, the main component of the standard bentonite, has a lot of permanent negative charge on the basal surface[37]. They are between 90–95% of the total charge[38]. As a result, the zeta potential of Bentonite drilling fluids is negative over the entire pH range.

Figure (9) displays the enhancement of zeta potential as a function of nanoparticles concentration for base water-based mud at room temperature. Results indicate that an increase in the nanoparticle Concentration enhances the zeta potential. The laboratory results display that the zeta potential of the water-based drilling fluid is improved by around 13% in the presence of 0.8 wt% Gr-Al_2O_3 at room temperature. The drilling fluid suspensions with high charge are appropriate during drilling operations to retain the suspension components discretely. The resultant drilling mud particles can penetrate to the porous wall of the wellbore and clog the pores. Also, a thin and impermeable cake is formed which minimize drilling fluid losses.

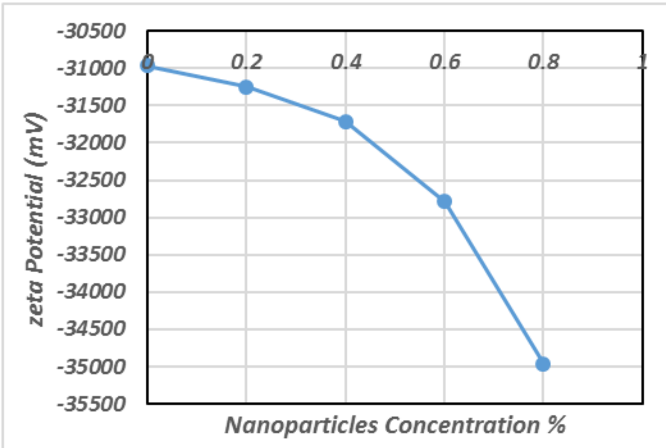

Figure 9. Zeta potential of prepared drilling fluids as a function of nanoparticles concentration.

Conclusions

During recent decades, researchers and scientists discovered nanoparticles, and currently, there are efforts to use this technology in the drilling operation. In this research, we explore the possibility of using hybrid nanoparticles to develop water-based drilling fluids properties. Based on experiential research that has been undertaken, it is possible to conclude that hybrid nanoparticles could be employed as multi-functional additive for drilling muds. In the present work, Gr-Al_2O_3 nanoparticles were applied as additives to water-based drilling fluids and below outcomes could be identified:

- Based on rheological investigations, the Nanoparticles addition has the power to form a drilling mud that displays sufficient features both at relaxation and in movement.
- The experimental outcomes demonstrate that the water-based drilling mud thermal conductivity is improved by around 10% in the presence of 0.8 wt% Gr-Al_2O_3 at room temperature.

- The laboratory results indicate that the water-based drilling mud electrical conductivity is improved by around 8.4% when the concentration of Gr-Al_2O_3 was 0.8 wt%.
- The laboratory outcomes identify that the water-based drilling fluid zeta potential is enhanced by around 13% in the presence of 0.8 wt% Gr-Al_2O_3.

In our future research, we intend to concentrate on the possibility to use different hybrid nanoparticles to enhance lubrication, filtration and other drilling fluid properties.

Acknowledgments

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