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Influence of Graphene Nanoplatelet and Silver Nanoparticle on the Rheological Properties of Water-Based Mud †

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Featured Application: Drilling fluid.

Abstract: Water-based mud is known as an environmental-friendly drilling fluid system. The formulation of water-based mud is designed to have specific rheological properties under specific oil field conditions. In this study, graphene nanoplatelet and silver nanoparticle (nanosilver) were added to a water-based mud formulation in which they act as drilling mud additives. Rheological properties measurements and filtration tests were conducted for evaluating the influence of the added nanoparticles. The results showed that the graphene nanoplatelet and the nanosilver increased the plastic viscosity (PV) by up to 89.2% and 64.2%, respectively. Meanwhile, both the yield point (YP) and the fluid loss values were reduced. In addition, we believe this is the first result ever report where nanosilver is utilized for enhancing-enhanced water-based mud's performance.

Keywords: graphene nanoplatelet; nanosilver; drilling mud; rheological properties; filtration test

1. Introduction

Drilling mud is known as the flowing fluid used during the drilling operation of oil and gas production. The main functions of drilling mud are to maintain the hydrostatic pressure, suspend and bring up the drilling solid cuttings to the surface, provide a lubricant and cool down the drill string and drill bit, and minimize formation damage [1,2]. As deeper and longer wells are drilled, the engineers face greater problems and challenges. The synthesizing and preparation of drilling muds must become more complex to satisfy the various operational demands and challenges that are attributed to formation characteristics. In particular, degradation of drilling mud additives, gelation or breakdown of polymeric additives during drilling operation can affect the performance of conventional water-based muds [3]. These drilling mud additives are chemicals added to drilling mud in order to change the mud properties and composition [4]. Nonetheless, many efforts have been devoted to drilling mud formulation mainly to enhance the quality and function of drilling muds, as well as to comply with the more stringent laws on underground or marine contamination.

Filtrate loss is another alarming issue during an extended-well drilling operation. It occurs in high permeability formation where the drilling mud seeps through the wall of the wellbore and invades the formation. The solid residue of the drilling mud will deposit a layer of mud cake on the wall of the borehole. Commonly, filtrate loss is controlled by the addition of a control additive, which are substances that can reduce the volume of the filtrate during propagation of a hydraulic fracture such as bentonite, starch, and organophillic lignite. However, bentonite degrades at high pressure-high temperature (HPHT) conditions.

Based on these notions, nanoparticles are being explored to improve drilling mud formulation in which they function both as weighing agent and filtrate loss control agent. The utilization of nanoparticles is of increasing interest to the oil industry with the intention of improving the performance of water-based drilling fluids as reflected by the efforts shown in Table 1.

Table 1. Recent research on nanotechnology in drilling mud system.

Nanoparticles	References
Multi-Walled Carbon Nanotubes	[5,6]
Graphene Nanoplatelet	[5]
Silica	[7–9]
Zinc Titanate	[10]
Zirconium (IV) Oxide	[11]
Chitin Nanocrystals	[12]
Titanium	[9]

It is reported the nanoparticles could improve drilling mud formulation; the main attributes are the particle size and the interaction potential with the formation, which might be due to the high availability of the active site. Al-Yasiri et al. [13] have reported a review on the role of drilling mud during the drilling process in oil and gas production, followed by the recent advances in the utilization of nanoparticles in a drilling fluid system. They summarized the impact of various types of nanoparticles on drilling mud performance to make the drilling operation smooth, cost effective, and efficient.

Abduo et al. [6] investigated the effect of carbon nanotubes and found that the rheological properties of water-based drilling mud were improved. They reported that carbon nanotubes can increase the shear stress; also, the shear stress was linearly proportional to the shearing rate. This is due to the better dispersion of drilling mud with the presence of carbon nanotubes at high shear rates. Also, the viscosity and the yield points were found to be higher for carbon nanotubes-added drilling mud than those of the conventional water-based mud. This means that the lifting capacity of the carbon nanotubes-added drilling mud is increased.

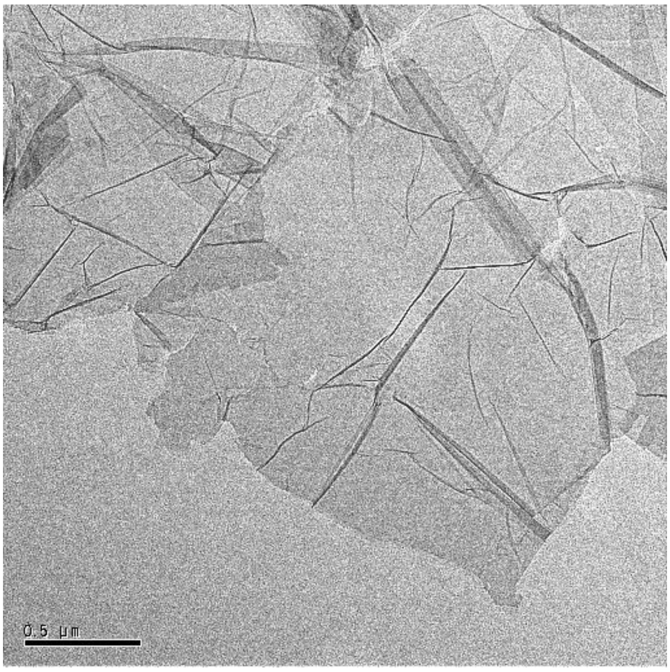
Ismail et al. [5] have experimentally investigated the effect of multi-walled carbon nanotubes and graphene nanoplatelets in water-based mud. They reported that the addition of 0.01 ppb of multi-walled carbon nanotubes and graphene nanoplatelet increased the plastic viscosity (PV) after a hot-roll of water-based mud from 21 cP to 23 cP and 25 cP, respectively. They analyzed the results and concluded that both multi-walled carbon nanotubes and graphene nanoplatelet increased the PV and reduced filtrate loss.

The features of nanoparticles in drilling mud formulation are further elucidated by utilizing the developed drilling mud at an oil field. Krishnan et al. [14] conducted an experiment on the effects of boron-based nanomaterials for drilling mud additives and it was used in an oil field in Myanmar; whilst Al Ruqeishi et al. [15] used carbon nanoparticles and zinc oxide nanowires to modify the water-based mud from the Wadi Laitham oil field.

The aim of this study is to investigate the performance of easy to maintain, economically feasible water-based mud in the presence of graphene nanoplatelet and nanosilver fluid additives. To date, there is no literature on nanosilver in a drilling fluid application. We believe this is the first result ever reported on the effect of nanosilver on the performance of water-based mud. In this study, the performance of water-based mud, including changes in density, rheological properties, and a filtration test, was evaluated. Rheological properties of drilling muds such as mud density, plastic viscosity (PV), yield point (YP), gel strength, and mud filtrate loss volume are important to maintain for an efficient drilling operation and wellbore stability. Note that this paper will exclude the discussion on the possible limitations or weaknesses of the nanoparticle in a drilling application such as the cost of nanoparticles, availability of the supplier and other factors due to a lack of existing literatures and we recommend this as future studies.

2. Materials and Methods

Graphene nanoplatelet was synthesized at Inha University, Korea via the modified Hummers method [16,17]. Figure 1a shows the distinctive layered structure of the graphene nanoplatelet obtained from exfoliated graphite identified by transmission electron microscope (TEM) (model: CM200, Philips, Amsterdam, The Netherlands). The crystalline structure of the graphene nanoplatelet was identified by X-ray diffraction (XRD) (model: DMAX-2500, Rigaku, The Woodlands, TX, USA) in Figure 1b displaying a peak at approximately 10.88°. Graphene nanoplatelet is colloidal suspended particles in aqueous solution produced by the chemical modification of graphite. Graphene nanoplatelet has multi-functional properties and it consists of 3–8 layers (average thickness), a diameter of particle ranging from sub-micron to 50 μm and the surface area is 750 m^2/g. It signifies the new class of nanocarbon. Graphene nanoplatelets can improve the matrix material such as the surface hardness, stiffness, and strength. Such attributes of graphene nanoplatelets are suitable for inhibiting clay swelling and wellbore strengthening. Nanosilver was supplied by Terra Techno Engineering, Shah Alam, Selangor, Malaysia. Its physical appearance is grey in color with an average size particle of ~50 nm and an average surface area of 3.7 m^2/g. Figure 2 shows the UV-visible absorption (UV-Vis) spectrum of the nanosilver particle showing a band at a range of 400–425 nm.



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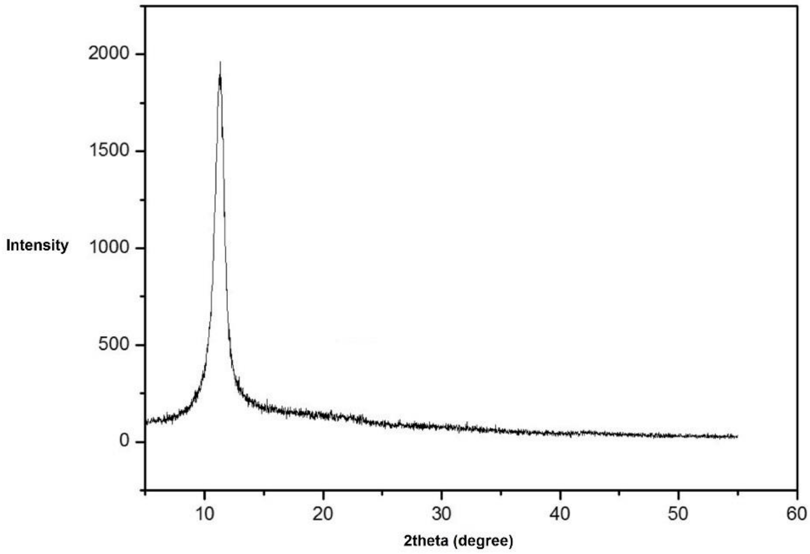

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Figure 1. (**a**) Transmission electron microscope (TEM) image; and (**b**) X-ray diffraction (XRD) spectrum of graphene nanoplatelet.

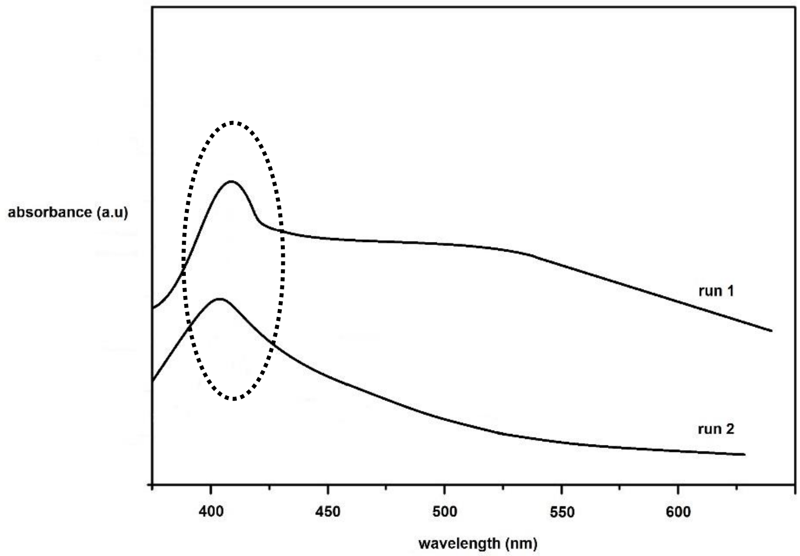


Figure 2. UV-Vis spectrum of nanosilver.

Table 2 shows a commercial formulation of water-based mud obtained from the Scomi Group Berhad company. The selected materials follow the ANSI/API 13A/ISO 13500 Specification for drilling mud materials standard. Eight samples of water-based mud were prepared using an electronic mixer (model: Hamilton Beach, Hamilton, OH, USA). Every time an addition of a component was made, a continuous stirring for about five minutes was performed. Step wisely, fresh water and pre-hydrated gel were added, followed by soda ash, potassium chloride, Hydrozan®, Hydro star NF® [18], and barite. Graphene nanoplatelet and nanosilver were added at different amounts as an additive to the prepared drilling mud samples. The mixture was stirred for about one hour to confirm homogeneity. All tests were conducted according to the ANSI/API 13B-1/ISO 10414-1 standard.

Table 2. Formulation of water-based mud.

Material	Function	Quantity (g)
Fresh Water	Base	287.22
Prehydrated Gel	Additive-viscosifier	22.56
Soda Ash	Additive-hardness control agent	0.20
Potassium chloride	Additive-ion exchange agent	39.13
Caustic Soda	Additive-pH control	0.70
Hydrozan®	Additive-viscosifier	1.50
Hydro star NF®	Additive-fluid loss agent	141.49
Barite	Additive-weighing agent	3.50

Density of water-based mud was measured using a mud balance equipment of accuracy to within 0.01 gcm^{-3}. The lid of the balance was removed from the balance cup and drilling fluid sample was placed inside the cup. The cup was covered with the lid and some drilling fluid was ensured to overflow from a hole of the lid to make sure the cup was fully filled with drilling fluid. Then, the arm balance was placed on the base, with the knife-edge resting in the fulcrum. The rider was moved along the graduated arm until the vial was centered. Density measurement was recorded at the left hand edge of the rider.

Rheological properties of water-based mud were determined by using a viscometer (model: Fann, Houston, TX, USA). The equipment was switched on and allowed to stabilize, after which the viscosity of distilled water was tested to check the integrity of the equipment. The mud sample was poured into the cup of the viscometer and was placed on the viscometer stand. The stand was adjusted and held in position as the rotor sleeve was immersed in the mud exactly to the fill line. The speed selector knob was selected to rotate at 600 rpm (revolutions per minute), and the power was switched on. When a steady dial reading was reached, this was recorded as the 600 rpm dial reading. The above process was repeated for 300 rpm. Consequently, plastic viscosity (PV) and yield point (YP) of the mud samples were calculated based on the two-data point approach using the equation:

$$\text{PV (cP)} = \theta_{600} - \theta_{300} \text{ reading}, \quad (1)$$

$$\text{YP (lb/100 ft}^2\text{)} = \theta_{300} \text{ reading} - \text{PV}, \quad (2)$$

where PV is plastic viscosity (unit cP), θ_{600} is dial reading at 600 rpm, θ_{300} is dial reading at 300 rpm and YP is yield point (unit lb/100 ft^2).

A filtration test was conducted at standard room temperature by using a high pressure, high temperature filter press equipment (HPHT filter press Series 387). Volume of filtrate of the sample was measured at a pressure of 500 psi, after the sample was left for 30 min. in a standard cell (no. 5 Whatman filter paper). The volume of filtrate loss was recorded from the graduated cylinder.

3. Results

Figure 3 shows that the presence of graphene nanoplatelet and nanosilver gave an insignificant effect on the density of water-based mud (mud weight). Density of the blank sample is 9.00 ppg (i.e., pounds per gallon). This is a common density used during the first stage of a drilling operation [19]. A key feature of mud density measurement is to reflect the ability of the mud to carry drilled cuttings as the suspending mud has an associated buoyancy effect on the cuttings. In the presence of graphene nanoplatelet and nanosilver at 0.1 g, 0.5 g and 1.0 g, the density increment is less than 0.50 ppg. In a drilling operation, mud density is designed slightly higher than formation pressure to ensure maximum penetration rate [19]. Too high mud weight will cause loss circulation of formation, stuck of drill pipe, decrease on penetration rate and formation damage. While insufficient of mud weight (too low) will cause well collapse or instability of the well due to formation pressure higher than the hydrostatic pressure provided by mud column. Ultimately, selecting mud density during the formulation stage is a key factor to realizing an effective drilling operation and remain economical. In general, the presence of graphene nanoplatelet and nanosilver gave an ambiguous effect on the mud weight due to the negligible dimension of its particle size. Mathematically, mud weight is calculated by the sum of weights over the sum of volumes. Since the weight of the added nanoparticle is negligible, the mud weight measurement will not be affected.

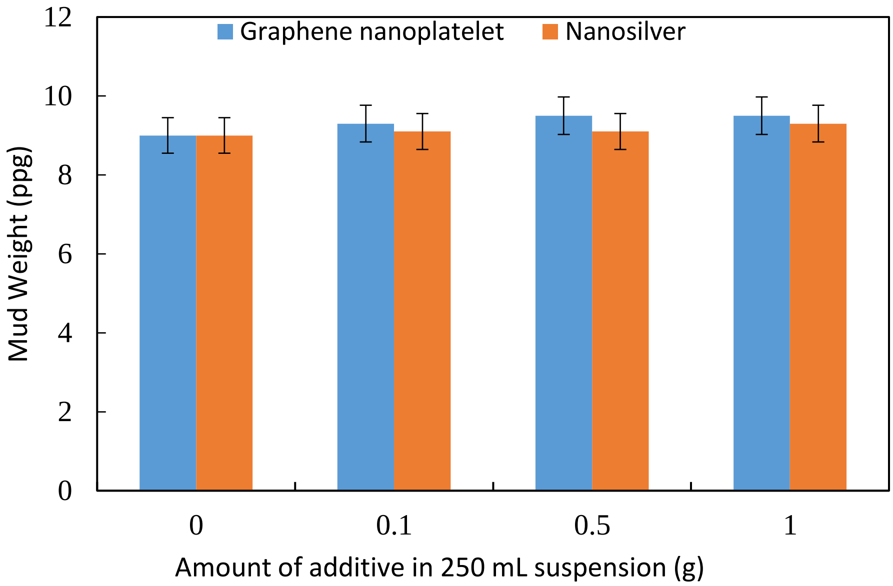
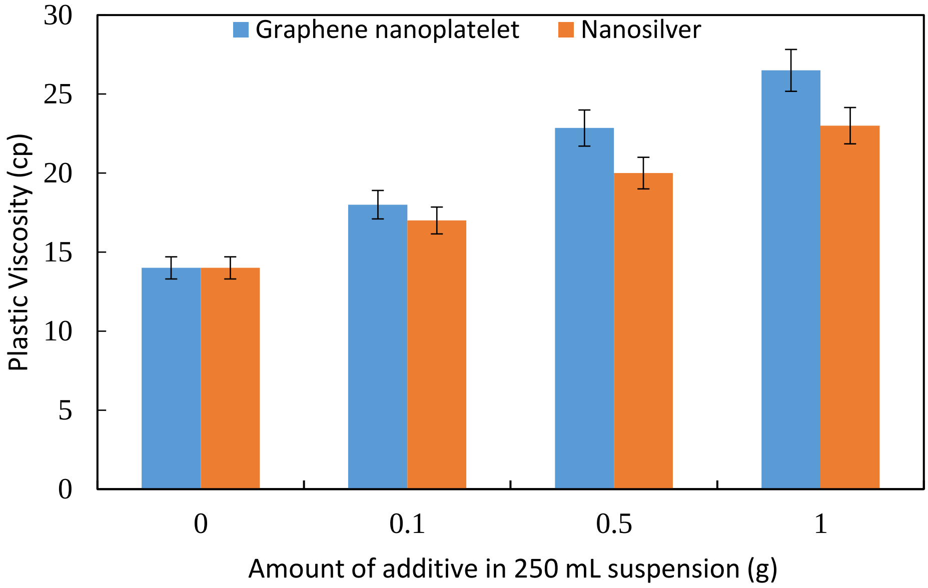


Figure 3. Density measurement of water-based muds with 5% error bar.

Plastic viscosity (PV) is defined as a resistance of fluid to flow [4]. It is characterized as a major aspect of the stream resistance of the liquid created by mechanical friction within the fluid. PV represents a viscosity of drilling mud. Figure 4a shows the viscosity of water-based muds with and without the presence of the nanoparticles. Significant increment due to the presence of a graphene nanoplatelet and nanosilver as shown in Figure 4a is reflected in Figure 4b, in terms of percentage.

Figure 4b shows that, with only 0.1 g of either the graphene nanoplatelet or the nanosilver, the increment of PV is up to 21.4% and 28.5%, respectively. At 0.5 g, the increment is tripled for graphene nanoplatelet and doubled for nanosilver. While at 1.0 g, both nanoparticles increased the mud viscosity to more than 50%. The statistical correlations for the mud viscosity in the presence of nanoparticle (NP) are (i) $PV = -0.3417\ NP^3 + 2.475NP^2 - 1.0333NP + 12.9$ (for graphene nanoplatelet) and $PV = 3NP + 11$ (for nanosilver) with both of the correlations having $R^2 = 1$. The presence of nanoparticles in the drilling mud formulation contributed to (i) high solid content, and (ii) high stability of waterborne particles. This is the reason why the PV measurement was observed to be increased without the formation of additional attractive force. The Dejaguin–Landau–Verwey–Overbeek (DLVO) theory is a basis for theoretical calculations explaining the nanoparticles behavior in a solution. According to the DLVO theory, the stability of particles is determined by the net electrostatic surface interactions of the particles and their Van der Waals forces. The net electrostatic surface interaction between the nanosilver particles is further investigated by measuring the yield point (YP) values.



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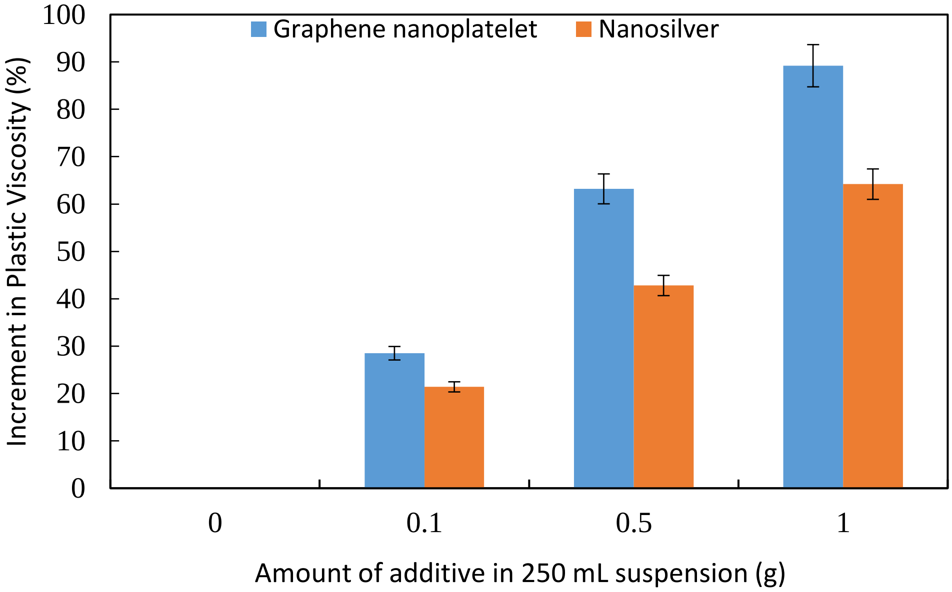

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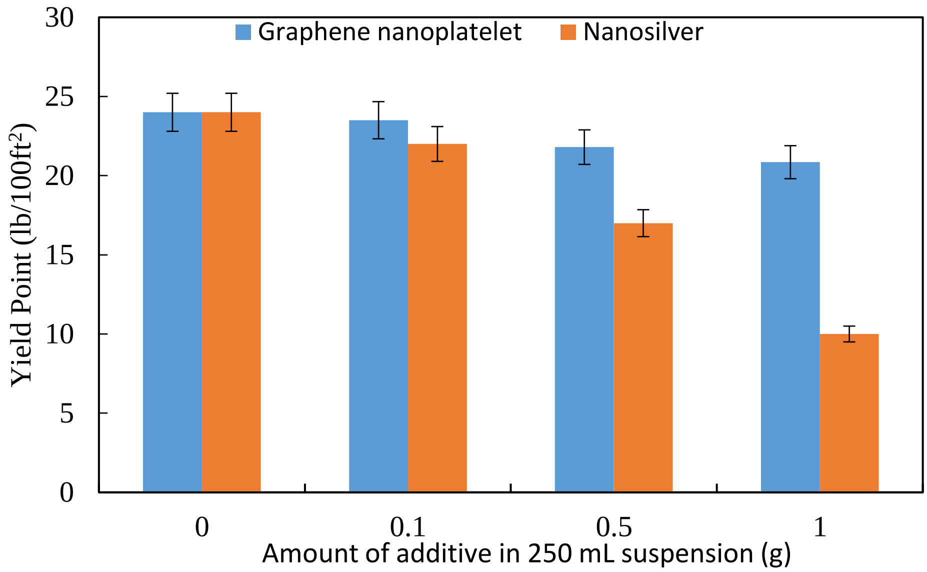
Figure 4. Plastic viscosity (PV) measurement of water-based muds with 5% error bar. (**a**) PV measurements, and (**b**) PV percent increment when graphene nanoplatelet and nanosilver are present.

YP is the initial resistance to flow caused by the electrochemical forces between the particles [20,21]. Technically, it indicates how much pump pressure must be applied during a drilling operation in order to bring rock cuttings up to the surface level from the static state. Meanwhile, in terms of surface interaction, high YP means an attractive surface force is exhibited and low YP means steric surface force is exhibited. When YP was determined as a function of a nanoparticle, a reduction of YP in the form of a 3rd order polynomial relationship was revealed, as shown in Figure 5a,b. The statistical correlations for the yield point measurement in the presence of nanoparticle (NP) are (i) $YP = 0.325NP^3 - 2.55NP^2 + 4.875NP + 21.35$ (for graphene nanoplatelet) and $YP = 0.1667NP^3 -$

$2.5NP^2 + 4.3333NP + 22$ (for nanosilver) with both of the correlations having $R^2 = 1$. Mud samples with graphene nanoplatelet added were not significantly affected. The percentage reduction is less than 14%. Meanwhile, the presence of 0.1 g nanosilver gave a YP reduction of 8.3%. At 0.5 g, the reduction is more than 3rd-magnitude (i.e., 29.1%). At 1.0 g, the percentage of reduction is in the magnitude of 5th (i.e., 58.3%).



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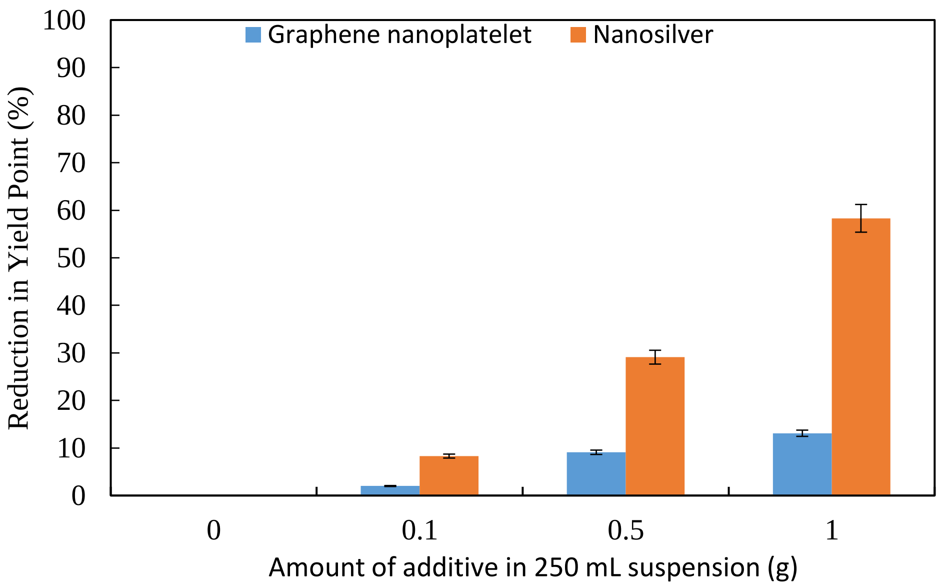

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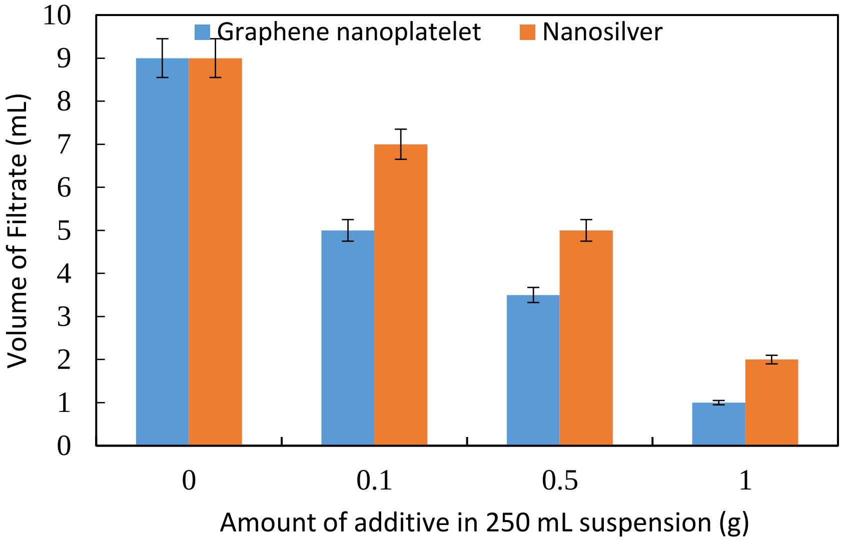
Figure 5. Yield point (YP) measurement of water-based muds with 5% error bar. (**a**) YP measurements, and (**b**) YP percent reduction when graphene nanoplatelet and nanosilver are present.

The effect of reduced YP revealed the existence of steric force formed under fixed ionic strength condition, between the nanoparticles in a colloidal water-based mud system. In general, water-based mud with a presence of nanoparticle has a lower YP as compared to a conventional type. This means that only low force or stress is needed for the drilling fluid to flow. It is indeed highly desirable to have a low YP to allow sand and shale cuttings to settle out and entrained gas to escape, to minimize swabbing effect during pulling the string out of hole, and permit the circulation to be started at a low pump pressure (i.e., economically feasible) [22]. In this study, we found that nanosilver is a potential candidate as it exhibits lower YP than that of graphene nanoplatelet. Previous studies have found that graphene compound acts as a surfactant, due to its capability in creating highly stable Pickering emulsions of organic solvents like solid particles [23]. It can also act as a molecular dispersing agent to process insoluble materials such as graphite and carbon nanotubes in water [24]. Conversely, the insignificant reduction of YP revealed in this study is likely due to particle bridging effect caused by the interaction of polar functional groups, located in an ideal position to interact with interacting adsorbed layers form nearby particles [17]. The presence of additional attractive force, such as a bridging interaction caused by adsorbed nanoparticles, was found to accentuate the bridging interactions which extent the flocculation state of suspension [25] and significantly increases the strength of interparticle interactions by up to 10-fold [26].

Figure 6a shows the performance of water-based mud in the presence of graphene nanoplatelet and nanosilver on the volume of filtrate (fluid loss); whilst, the percentage reduction is shown in Figure 6b. The filtration test was conducted at 500 psi and it is within an acceptable range of hydrostatic pressure (200–500 psi). In designing drilling mud, the hydrostatic pressure to be imposed by the drilling mud must be higher than the formation pressure. If the drilling mud was incorrectly formulated, the expected hydrostatic pressure may not be attained. Hence it will initiate the influx of formation fluid to occur and a phenomenon that is known as 'kick' will be triggered and may result in blowout. Generally, a blowout will occur when a well kick could not be controlled or 'killed'.

Figure 6c shows the odourless, colourless filtrate collected in this study. Without the presence of nanoparticle, the fluid loss was 9 mL. At 0.1 g, 0.5 g, and 1.0 g graphene nanoplatelet, the fluid loss was 5.0 mL, 3.5 mL, and 1.0 mL, respectively. When nanosilver was added to the sample with the same amount, 7.0 mL, 5.0 mL, and 2.0 mL of filtrate was collected, respectively. It is shown that the addition of nanoparticle has successfully reduced the filtration loss up to 89% at ambient temperature. This is attributed to the fact that the nanoparticles have been completely emulsified in the mud samples, thereby forming a stable mud suspension. Also, results showed that the graphene nanoplatelet performed more efficient than nanosilver. Formulation of drilling mud with low volume of filtrate is highly desirable when designing drilling mud system. In a drilling operation, high fluid loss can cause the invasion of drilling muds and hence borehole instability could reduce the reservoir potential and the amount of productivity [27]. Nonetheless, it is observed that nanoparticles show potential as fluid loss additives and a filtration agent for water-based mud.

The texture of mud filter cake is one of the parameters to be concerned in evaluating drilling mud performance. The mud filter cake characteristics [28] such as smooth or coarse and the amount of voids and pits can be attributed to the type of additive added to mud formulation. It was observed that the mud filter cake with added nanoparticle, as shown in Figure 6d, was similar to that of the mud without nanoparticle. This might be attributed to the nanoparticle high surface to volume ratio. In designing a mud formulation, it is of great importance in making sure the mud filter cake developed from the drilling mud is thin, smooth and impermeable to prevent a drill bit sticking problem and formation damage while drilling [29,30].



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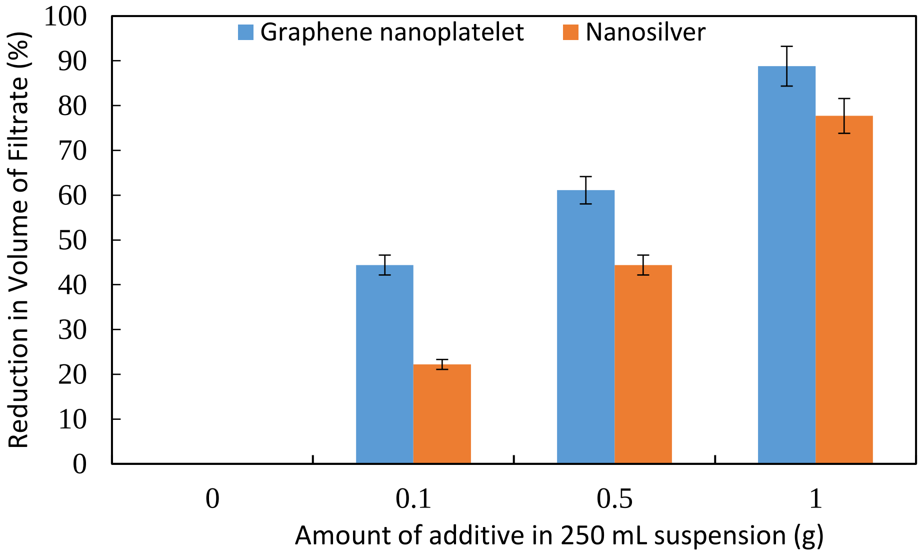

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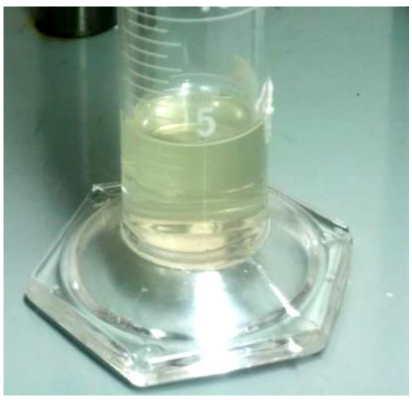

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Figure 6. Fluid loss measurement of water-based muds with 5% error bar. (**a**) Fluid loss measurements, (**b**) Fluid loss percent reduction when graphene nanoplatelet and nanosilver are present, (**c**) Filtrate collected from the HPHT filter press, and (**d**) Mud filter cake produced at 500 psi.

4. Conclusions

Graphene nanoplatelet and nanosilver were utilized to enhance water-based drilling mud properties. According to density, rheology, and filtration studies, both graphene nanoplatelet and nanosilver are potential candidates as a chemical additive in the formulation of water-based mud. In this study, the results showed that the presence of graphene nanoplatelet and nanosilver gave insignificant effect on mud weight (density). The drilling mud with added graphene nanoplatelet exhibits an increment of its plastic viscosity by up to 89.2%. Unlike the nanographene platelet, the nanosilver increased the mud plastic viscosity by only 64.2%. Both graphene nanoplatelet and nanosilver reduced the yield point by 13.1% and 58.3%, respectively; in the form of 3rd order polynomial correlation. A similar effect is observed with the fluid loss measurement (volume of filtrate) where the graphene nanoplatelet and nanosilver reduced the fluid loss by 89.0% and 77.7%, respectively. We also found that both the mud filter cake without the presence of nanoparticles and the mud filter cake with added graphene nanoplatelet or nanosilver are similar in which the texture is thin and smooth. In the future, these nanoparticles will be utilized at elevated temperatures and pressures for water-based drilling mud's performance enhancement.

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Conflicts of Interest: The authors declare no conflict of interest.

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