Research Article

Study on the Effect of Nanoparticle Loadings in Base Fluids for Improvement of Drilling Fluid Properties

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Abstract

Nanotechnology is increasingly capturing the attention of material researchers as this technology pushes the limits and boundaries of the pure material itself. Liquids dispersed with nanoparticles generally have higher physical properties enhancements. In the current study, ball-milled functionalized –COOH carbon nanoparticles were introduced into the targeted base fluid of drilling mud. The investigating parameter involved in this study is carbon nanoparticle loadings, ranging from 0 wt% to 1.0 wt%, which is readily dispersed in the base fluid. The method of dispersion chosen is through indirect dispersion in ultrasonic bath. The effect and significance of the investigating parameters were studied based on the desired physical properties of ideal base fluids for drilling muds, mainly thermal conductivities and viscosity of the fluid. The conditions for dispersion in this study were ball-milled functionalized –COOH carbon nanoparticle size at an average size of 10 μm with 90 minutes of indirect ultrasonic dispersion. Result shows that addition of functionalized nanoparticles into base fluids yields as much as 6% of thermal conductivity enhancement while approaching viscosity of pure base fluid at higher shear rate.

Keywords: Nanoparticle; Ultrasonic dispersion; Drilling fluid; Thermal conductivity; Viscosity

Introduction

Drilling fluid, commonly known as drilling mud, is one of the most important aspects in any drilling process. It primarily aids to cool and lubricate the drilling bit, remove solid fragments from drilling area to the surface and counterbalancing the formation pressure within the wellbore.

However, the exploration of oil and gas gradually ventures deeper and further into deep sea drilling when high volume of oil and natural gas can be found. Deep sea drilling is often conducted in a pre-set environment at high temperature high pressure (HTHP) due to frictional forces and pressure due to extreme depth of the wellbore. Commonly, this poses mechanical malfunctions to drilling equipment used. Such problems include overheating of equipment as well as lost circulation is common in any drilling operations due to the limitations posed by drilling mud.

In recent years, nanotechnology is starting to make its impact globally by expanding the current limitations of current available materials and fluids. The properties of materials are differentiated from the conventional material used. Nanomaterials have a relatively larger surface area. Thus, this enhances the material as they are chemically more active and enhancement in terms of strength, electrical and thermal properties can be seen [1]. The integration of nanotechnology into oil and gas exploration sector opens up new opportunities to further explore the capabilities of oil and gas exploration in deep water.

Dispersion of nanoparticles is commonly carried out using high intensity ultrasound. However, various parameters generally affect the dispersion of dry powdered nanoparticles, mainly particle adhesion/cohesion forces [2], surface tension of fluids [3], and nanoparticle size [4,5].

Nanofluids generally produce higher thermal properties compared to its counterpart base fluid [3,4]. However, the viscosity parameter has significant fluctuating behaviours as there is sharp increment of nanofluid viscosity before approaching base fluid's viscosity [4].

In this study, the experimental design setup for dispersion of nanoparticles involves preparation of carbon nanoparticles beforehand. Ball-milling of carbon nanotubes is carried out to ensure reduction of size of tubular carbon tubes at nanoscale. Nanoparticle loadings considered in this research study are in accordance to the range carried out by Sedaghatzadeh, et al. [5] at 0.2 wt%, 0.4 wt%, 0.6 wt%, 0.8 wt% and 1.0 wt% ratio of nanoparticle mass to mass of base fluid. Functionalised carbon nanoparticles are dispersed into oil-based mud, which is a non-polar liquid. The dispersion of hydrophobic nanoparticles into a non-polar fluid is suitable as it more stable and more soluble due to its counterparts. Previous studies have been conducted on hydrophobic nanoparticles dispersed in oil-based fluid are able to obtain excellent dispersion process through additional adjustments in pH and surface-capping of nanoparticles in the nanofluid suspension [6]. Functionalized nanoparticles are able to stabilize itself well with stability up to six (6) months without sedimentations [7]. Besides that, influences of ball-milling, temperature and ultra-sonication intensity are considered in this study.

This article aims to portray that the addition of minimum amount of functionalized carbon nanoparticles are able to yield enhancement in thermal conductivity as well as retaining the viscosity of the nanofluid

suspension as it approaches higher shear rate. This effect is particularly important in any drilling operations as higher thermal conductivity of drilling fluids are required to channel heat load away from the drilling bits to avoid equipment overheating. Besides that, viscosity is an important parameter as addition of functionalized carbon nanoparticles gives higher viscosity at low shear rate while it approaches viscosity of the base fluid at higher shear rate. This effect is beneficial to any drilling operations as it shows that less power consumption is actually needed for drilling operations which utilizes drilling nano-fluids.

Materials and Methods

Materials

Functionalized carbon nanotubes are selected in this research study. The carbon nanotubes have an average size of 24 μm. A particle size distribution (PSD) analysis is provided in Figures 1 and 2.

Nanoparticles preparation

Planetary Mono Mill PULVERISITTE 6 *classic line* series is used in the ball milling process. The cylindrical bowl with zirconium oxide material of construction with 250 mL volume containing 25 zirconium oxide balls with mean diameter of 5 mm are filled with 20 g of carbon nanotubes. The functionalized carbon nanotubes are ball-milled into carbon nanoparticles through ball-milling operation. The approach taken in this preparation follows Tucho, *et al.* [8]. The total ball-milling duration is three (3) hours with intensity of 500 rpm. In this study, each hour of ball milling duration has an allowance of 10 minutes stoppage time and is repeated until total of 3 hours of ball milling duration is achieved.

Nanoparticles characterization

Two traits are characterised in this study approach, mainly the particle size distribution of the nanoparticles and the chemical bonds and functional groups present in the carbon nanoparticles.

The particle size distribution (PSD) analysis is conducted using Malvern Mastersizer 2000 Particle Size Analyzer, where it is used to measure the particle size where the scattered light intensity from the passing laser beam through the dispersed particulate sample is measured.

Fourier Transform Infrared (FTIR) spectroscopy analysis is carried out using Perkin Elmer Transform Infrared Spectrometer where transmittance of light at specific wavelengths is measured. The characteristics of the peaks recorded are classified into type of peaks, chemical bonds and functional groups present.

Base fluid and nanofluid characterization

The thermal conductivity and density are measured in this study.

For density measurement, Anton Paar Density Meter 4500 M is used. A 10 mL of pure base fluid is injected into the side port of the device. The method of measurement is through the differences of fluid in U-tube located within the device. The reference temperature of the measurement is taken at 25°C.

For thermal conductivity analysis, P.A. Hilton Thermal Conductivity of Liquids and Gases Unit H471 is used. The heater is jacketed by a plug with two thermocouples positioned in two different locations: at the plug and at the jacket. The thermal conductivity of base fluid is measured by determining the temperature difference between two thermocouples across a fixed heat transfer area. The radial clearance used in this research is 0.3 mm with 0.0134 m² heat transfer area. The voltage applied to the heater is constant at 60 V for liquid while 40 V is applied for gas. The formula of conductivity used in this analysis is shown below:

$$\text{Thermal conductivity, } k = \frac{Q\Delta r}{A\Delta T} \qquad (1)$$

where Q is the heat transfer by conduction, W; Δr is the radial clearance, m; A is the surface area of heat conduction, m²; and ΔT is the temperature difference between two thermocouple points, °C.

The incidental heat transfer present must not be neglected when performing calculations of thermal conductivity of the base fluid. The experiment was conducted twice and the average mean values were taken and plotted.

The viscosity analysis of the base fluid and nano-based fluid is carried out using Brookfield Cap 2000 + L series which has a supported temperature range from 5°C to 75°C. In this experimental study, the shear rate, in rpm, is carried out at 100, 150 and 200 respectively at temperature of 30°C. The spindle used in this study is spindle 3, with a running time of 60 s and holding time of 10 s. The experiment was repeated twice with the same spindle number and the average mean value was taken and plotted.
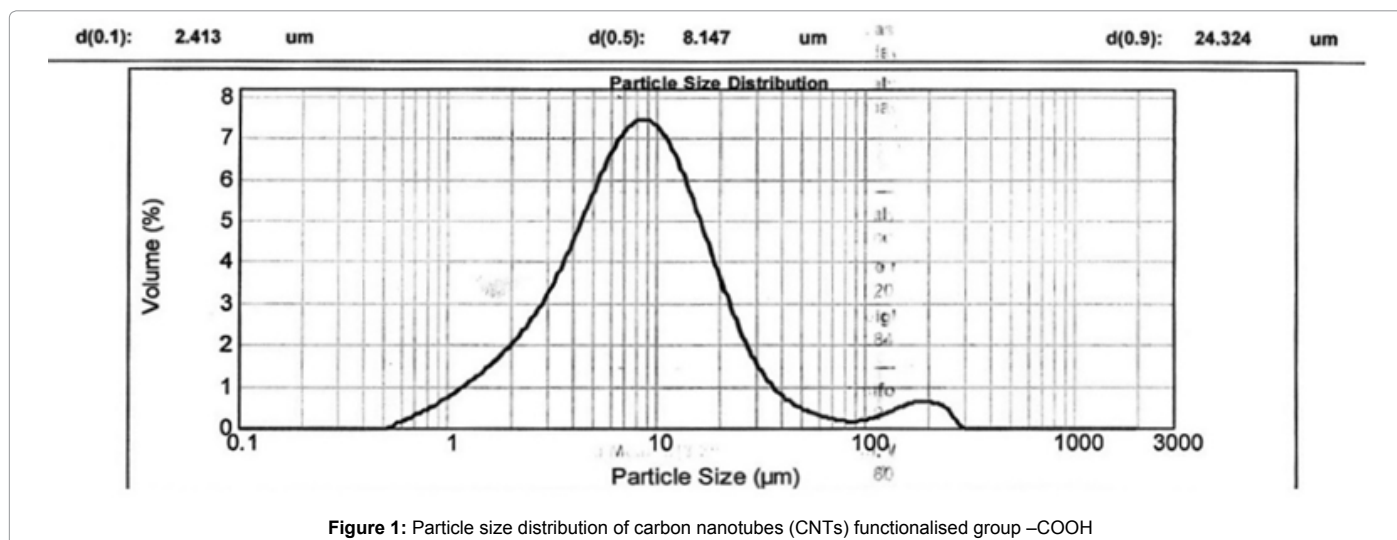

| d(0.1): | 2.413 | um | d(0.5): | 8.147 | um | d(0.9): | 24.324 | um |

Figure 1: Particle size distribution of carbon nanotubes (CNTs) functionalised group –COOH

Nanoparticles dispersion

In this research study, indirect sonication dispersion is carried out to disperse carbon nanoparticles into the base fluids. Bath Ultrasonic Branson Model 8510E-DTH equipment is used to suspend samples within the tank. The tank has dimensions of 495×290×150 which allows multiple samples to be sonicated together at the same time. The delivering power of the equipment is 320W with a frequency of 40 kHz. Five (5) samples of base fluid measuring 100 mL is mixed crudely with 0.2wt%, 0.4wt%, 0.6wt%, 0.8wt% and 1.0wt% of carbon nanoparticles respectively in 150 mL beakers. The carbon nanoparticle mass loadings are calculated beforehand based on the density of pure base fluid. The correlation used to estimate ultrasonic dispersion follows Yang et al. [9]. The correlation is as stated below:

$$E = \frac{P \times t}{V} \qquad (2)$$

where P is the output power of the sonicator, W; t is the sonication time, in s; V is the total volume of liquid sonicated, in mL. The optimum sonication energy estimated by Yang et al. is averagely 2250 ± 250 J/mL. From Equation 2, the time taken required to disperse nanoparticles with 100 mL of base fluid with constant power deliverance of 320 W is roughly 13 minutes. However, the dispersion of samples was also carried out at 15 minutes, 30 minutes and 90 minutes respectively.

Results and Discussions

Particle size distribution (PSD) analysis

The analysis of the particle size distribution is provided in the figures below. Figure 1 and Figure 2 display the results of CNTs before ball-milling and after ball-milling respectively. In Figure 1, 10% of the total population measured is averagely 2.413 μm and 90% of the total population giving an average measurement of 24.324 μm. In Figure 2, 10% of the total population gives a reduction in measurement length at 1.236 μm while 90% of the total population also gives a reduction in length at 12.273 μm.

Overall, the comparison in size of carbon nanoparticles before and after ball milling shows a total reduction in size by 48.8% with more or less reduction size of half of the length of the carbon nanotubes.

For the preparation of ultrasonic dispersion of nanoparticles, the pre-requisite for a good dispersion and a stable suspension requires nanoparticles at 200 nm or shorter. This is further supported by literature reviews of previous experimental works which dispersed similar sizes of nanoparticles. In this experiment, the ball-milled samples do not fully fit the requirement as 90% of the total population distribution yields sizes larger than 200 nm. The nature of the size of the carbon nanoparticles may give rise to the tendency of the carbon nanoparticles to agglomerate or tangled each other when dispersed under ultrasonic dispersion.

Ball milling of carbon nanotubes experiment is carried in accordance to the study conducted by Tucho et al. (2010) [8], where key parameters involved in this experiment are the speed factor and time duration for the ball milling process. 500 rpm and 3 hours of total duration of intense ball milling experiment were carried out. However, from previous experience the high intensity of the impact between the grinding balls and the walls of the grinding bowl might scrape off trace amounts of zirconium oxide into the samples.

Therefore, to prevent any contamination of zirconium oxide into the carbon nanoparticle samples, at every hour of ball milling operations, there will be an hour of interval stoppage before the next hour of ball milling. At the same time, this could also prevent deformation of the morphology of the carbon nanoparticles due to high temperature as a result of extreme intensity from the collision of the grinding bowls.

The collision intensity of the grinding balls might not build up sufficiently over the total 3 hours of ball milling duration as suggested by Tucho et al. (2010) [8] as there is an interval between each hour of ball milling operations.

Fourier transform infrared (FTIR) spectrometry

Figure 3 below shows the Fourier Transform Infrared Spectroscopy (FTIR) analysis result of ball milled carbon nanoparticles functionalised group –COOH. A table of characteristic IR is used to determine the presence of the bonds and functional group in the samples.

In Figure 3, a broad peak with wavelength 3428.7 cm⁻¹ can be seen which shows presence of O-H stretch bond or H-bonds which shows the presence of possible alcohol or phenol groups. A small peak can be seen at wavelength 2917.58 cm⁻¹ showing the presence of O-H stretches with carboxylic acids as its main functional group. The analysis shows medium peak at 1635.13 cm⁻¹ containing carboxylic acid functional group as well. Another smaller broader peak at 1130.35 cm⁻¹ also shows C-O stretch bonds containing carboxylic acid group. From the FTIR results obtained it can be concluded that no external organic
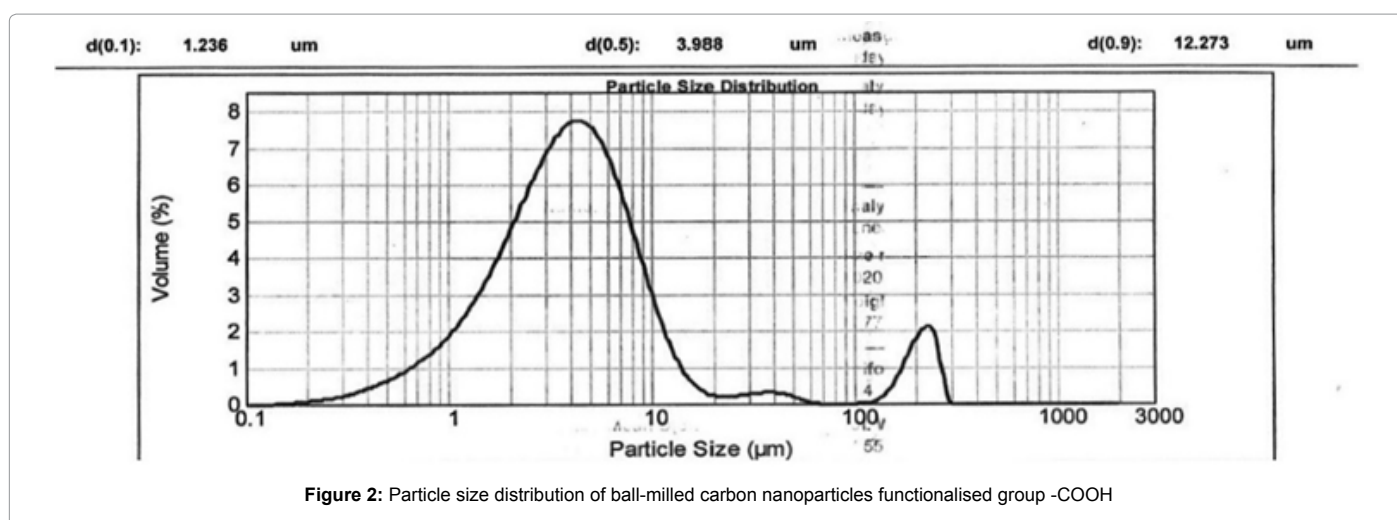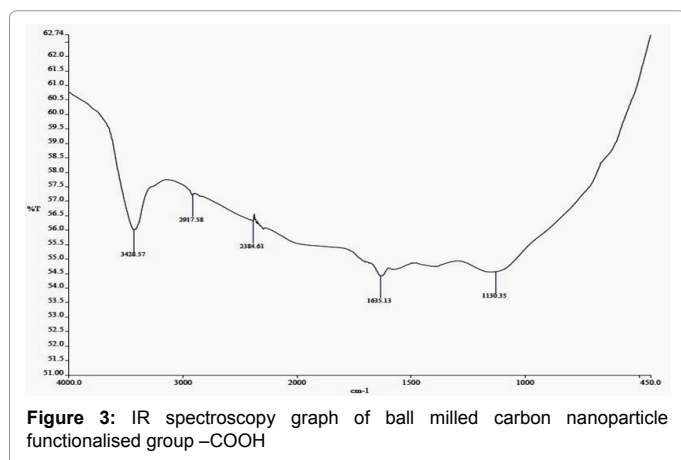

Figure 2: Particle size distribution of ball-milled carbon nanoparticles functionalised group -COOH

Figure 3: IR spectroscopy graph of ball milled carbon nanoparticle functionalised group –COOH

contaminants can be traced in this carbon nanoparticle sample.

Thermal conductivity analysis

In this experiment, the thermal conductivity of nano-basefluid is measured at temperature of 60 V settings. The temperature at the maximum voltage delivered is at mean temperature of 43°C. The thermal conductivity enhancement of the nanofluid can be summarised in Figure 4.

From Figure 4, a fluctuating trend can be seen when nanoparticle loadings are increased from 0.2 wt% to 0.6 wt%. Thermal conductivity of nanofluid increases nonlinearly with increasing mass fraction of nanoparticles [8]. This is true from the results obtained where the profile of relative thermal conductivity enhancement against nanoparticle loadings (wt%) increased nonlinearly. However, increasing nanoparticles concentration decreases the distance between particles while random collision due to Brownian motion from the heating process accelerates agglomerations of nanoparticles. The size of nanoparticles plays an important factor in determining the nanofluid's thermal conductivity. The ball milled nanoparticles in this study yield nanoparticle sizes of 4 μm averagely. However, large particles do not possess Brownian motion anymore as the particles approach micrometre size [9], thus leading to lower thermal conductivity enhancements. From the results obtained, 0.2 wt% has a considerable high relative thermal conductivity than 0.4 wt% and 0.6 wt% respectively due to increase distance between particles. However, higher nanoparticle loadings gives greater thermal conductivity enhancement but induced higher settlement of nanoparticle cluster sizes in the end.

Another possible cause for the fluctuating trends can be attributed to the inconsistency of water flow around the jacketed area of the equipment. Higher water flow rate cools down the shell side's temperature, resulting in higher difference of temperature gradient between both thermocouple points at the heater and shell side respectively.

Viscosity analysis

The analysis is conducted at a fixed temperature of 30°C to compare the viscosity of the pure base fluid with nanofluid samples. From the plot above, the pure base fluid has the lowest viscosity while 0.4 wt% of dispersed nanoparticle in base fluid has the highest viscosity at shear rate of 200 rpm.

The viscosity of the nanofluid suspensions has been widely debated where numerous studies found that there had been fluctuating results

from various researchers which can hardly be concluded [4]. The result shown by the plot shows that although it is true all nanofluids of various nanoparticle loadings have higher viscosity compared to its base fluid, the trendline shows that at higher shear rate, the viscosity of the nanofluid approaches that of the base fluids'. From Figure 5, the nanoparticle concentrations of 0.4 wt% are just slightly higher than that of 1.0 wt%. This can be attributed to the uneven dispersion of nanoparticles. A larger nanoparticle size will coagulate more easily to form larger agglomerates as larger size of nanoparticles cause decrement in distance between particles and increases the attractive van der Waals forces. Thus, nanoparticles at the point of extraction for sample 0.4 wt% are generally larger in size which attributes to the higher viscosity of nanofluid. As comparison, 0.4 wt% nanoparticle loadings also yield the lowest thermal conductivity as compared to the remaining loadings. It can be concluded that 0.4 wt% nanofluid sample has agglomerated sufficiently to yield lower thermal conductivity but higher viscosity [10].

Conclusions

The study on effects of nanoparticle dispersion loadings in base fluids is relative new research discoveries, in which limited research experimental data are available. The addition of nanoparticles into base fluids shows thermal conductivity enhancements at higher loadings. However, the size of nanoparticles plays a crucial role in affecting the physical properties of the base fluids.

Recommendations

Use of surfactants to suspend nanoparticles within the base fluid is recommended in future study. Study shows that usage of surfactants gives longer suspension duration of nanoparticles, resulting in better dispersion of the nanoparticles.
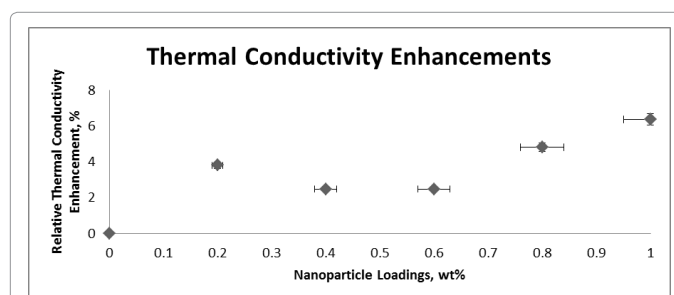
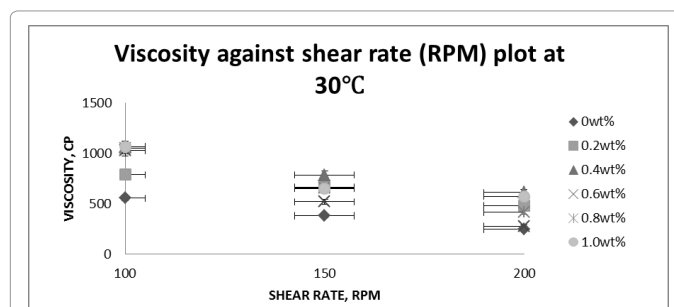**Figure 4:** Thermal conductivity enhancement as a function of nanoparticle loadings

Figure 5: Viscosity against shear rate, rpm, and plot at 30°C

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