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ORIGINAL ARTICLE

Improving the fluid loss and rheological characteristics of Oil-based drilling fluid by using Copper Oxide nanoparticles

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Abstract

A successful drilling operation depends strongly on a useful drilling fluid system. Using nanoparticles (NP)s to formulate intelligent drilling fluids gives them a wide range of optimal properties under different operating conditions and resolves any operational problems. In this study, in order to provide an effective solution for improving the rheological and high pressure/high temperature (HPHT) filtration properties of the oil-based drilling fluid (OBDF), copper oxide (CuO) NPs were synthesized with dandelion morphology. CuO dandelions were characterized by using XRD, FTIR, SEM, and zeta potential measurements. A long time stabilized nanofluid (NF) was prepared and evaluated. The OBDF samples consisting of various amounts of NF ranging from 1 to 11% (V/V) were prepared. Then, the fluid loss and rheological properties of OBDF were examined. The results showed that the OBDF containing 7% (V/V) NF was appropriate to improve the rheological properties such as yield point (YP), apparent viscosity (AV), and gel strength (GS). In addition, the minimum HPHT filtration value of 2 ml was acquired for the drilling fluid containing 9% (V/V) NF. In conclusion, CuO NPs demonstrated a positive effect on the performance of the OBDF system.

Keywords: *Dandelion Morphology, HPHT Filtration, Nanofluid, Viscosity, Yield Point.*

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INTRODUCTION

Drilling fluid, also known as drilling mud, is used to drill oil and gas wells. Drilling fluids are an essential element in the drilling process because of offering the mobility of drilling cuttings from the the bottom to the surface, providing buoyancy, its aids in drilling operations by cooling, cleaning and lubricating of bits, its pressure control and guarantee of the hole stability [1, 2]. The most commonly used drilling fluids include water-based drilling fluids, synthetic-based drilling fluids and OBDFs [3]. Although water-based drilling fluids are less expensive, non-toxic and more environmentally friendly [4]; but, in complex and more challenging drilling formations, OBDFs are preferred due to their potential to withstand a high pressure and temperature conditions, easy cleaning, high lubricity, significant shale inhibition [5].

OBDF is a liquid-liquid two phase system of dispersed water droplets in a continuous oil phase which is stabilized by emulsifiers [6]. Hence, the OBDF is also called invert oil emulsion. Rheological properties, as key characteristics of drilling fluid, play an important role in the drilling performance of oil and gas wells. The viscosity of the drilling fluid must be high enough to lift and remove drilling cuttings from the bottom of the hole. In addition to the rheology, fluid loss is the next property of drilling fluids that must be minimized to perform cheaper and safer drilling operations. Optimal reduction of fluid loss leads to the creation of an impermeable filter cake that minimizes the formation damage [7].

The various additives are added to drilling

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fluid to achieve the desired rheological properties, well stability and acceptable drilling operation. Selecting the type and amount of additives relies on the type of reservoir and the drilling method used. The major drilling fluids additives are viscosity modifiers, weighting materials, lubricants, filtration control additives, defoamers, thinning agents and emulsifiers [2, 8, 9]. Since additives have a great influence on the performance of drilling fluids, it is important to introduce a drilling fluid additive that results in good rheological properties and the lowest fluid loss.

With recent developments in NP technology, several studies have examined the possibility of using NPs in drilling operations [10-14]. The high surface area to volume ratio of NPs, along with their small size, makes NPs the most promising materials for the formulation and development of nano-based drilling fluids [3]. NPs can be effective in reducing fluid loss, shale inhibition, filling pores in the porous media and improving the thermal conductivity of drilling fluids [15-17]. In addition, particularly in the OBDF, nanomaterials can improve the rheological properties and thermal stability of drilling fluids under HPHT conditions [18]. Various nanomaterials have been used as additives in drilling fluid formulation such as nanosilica [19, 20], multi-walled carbon nanotube [17], nanopolymers [21], laponite [22], nano-graphene [23] and TiO_2/Polyacrylamide (PAM) [24]. Among these, metal oxide NPs are the most useful ones that are used by different drilling fluids. These NPs can stabilize and modify the rheological properties of drilling fluids, which improve the hole cleaning and prevent its instability [25, 26]. CuO NPs are one of the valuable metal oxides, which have remarkable properties including outstanding ductility, toughness, high hardness and great electrical conductivity. They are also semiconductors in nature that resulted to use them in devices such as electrochemical cell, catalysts, gas sensors and nano fluid [27, 28].

Various researches have been done on the possibility of using different additives in the drilling fluid formulations [29-31]. Although several studies have been performed on water-based drilling fluid additives, there is insufficient information on additives for OBDFs. The results of most scientific research are not commercialized due to some problems such as shortage of raw materials or high production costs of NPs. In this study, due to the mentioned properties of CuO NPs and because inexpensive raw materials required for the synthesis of CuO NPs are available in Iran, CuO NPs were synthesized and characterized. Then a long term stabilized NF containing CuO NPs were prepared and added to the OBDF as an additive to improve the HPHT filtration and rheological properties.

MATERIALS AND METHODS

Materials

The chemicals used in the synthesis of NPs and NF include urea, copper nitrate, ethylene glycol, distilled water, sodium hydroxide and sodium dodecyl sulphate. All the chemicals used in this study were purchased from Merck Company.

Instrument

X-ray diffraction (XRD) (Philips Co., Netherlands, model PW1730), Fourier Transform Infrared Spectrometer (FTIR) (model Avatar, Thermo Co., USA), scanning electron microscope (SEM) (model Mira 3-XMU, Tescan Co, Brno, Czech Republic), rotary viscometer (VG meter model 35A, Fann Instrument Co., Houston, Texas) and Filter press (HPHT Filter press 175 ml, Fann Instrument Co., Houston, Texas).

Synthesis of CuO NPs

Chemical reactions that used to take a long time to complete can now be done in minutes with the aid of a microwave. Microwave based synthesis has not only helped in implementing green chemistry, but has also revolutionized organic synthesis. Microwave irradiation is known to promote the synthesis of a variety of compounds, where chemical reactions are accelerated by the selective absorption of microwaves by polar molecules. Thus, it has been found that the main advantages of microwave irradiation synthesis methods over conventional methods are that they are fast, mild, energy-efficient and friendly to the environment. [32]. In this study, to synthesize CuO via microwave method, initially 0.16 g $Cu(NO3)_2$ added to 50 ml distilled water (I) and then 0.12 g urea (as a fuel for carrying out the combustion synthesis) was added to 20 ml distilled water (II) [27]. solution (II) was then added to the solution (I) and was irradiated in a microwave at 600 W for 1 h. Then, the resulting precipitate was centrifuged and placed in an oven at 140 °C for 1 h.

Preparation of NF

The synthesized CuO NPs must be dispersed initially to perform optimally as a drilling fluid additive [1]. Thus, the NF was prepared to stabilize dispersed CuO NPs and prevent particle agglomerating and precipitating. Among 43 NF samples (Data are not shown) which were prepared based on the design of experiment, a sample with the longest stability time (over 100 days) was selected for subsequent experiments. The specification of the selected NF is shown in Table 1.

OBDF Formulation

The specification of the OBDF as the base fluid is reported in Table 2. The formulation of the nano OBDF samples consists of various amount of NF, as shown in Table 3. The NF was used in the OBDF formulation by adding 1 to 11% (V/V) to calcium chloride brine. The long time stabilized NF was initially added to the aqueous phase of the OBDF (calcium chloride brine). The aqueous phase was then added to the oil phase (including gas oil, fluid loss control (FLC), lime, and primary emulsifier). After that, secondary emulsifier, oil-based viscosifier and limestone were added to the OBDF emulsion, respectively. The prepared nano OBDF was sufficiently heated and mixed. The rheological properties and HPHT filtration of nano OBDF samples were measured. The performance of the nano OBDF samples was compared with the base fluid.

Measurement of the rheological properties

The rheological properties of OBDF were measured at standard atmospheric pressure (0.1

Table 1. Specifications of the selected NF for using in the OBDF formulation.

	Morphology of CuO NPs	**EG /Water ratio (%)**	**Ultrasound time [hr]**	**CuO [wt%]**	**Sodium dodecyl sulfate (SDS) [wt%]**	**pH**
NF	Nano Dandelion	75	6	0.22	0.25	8

Table 2. The specifications of the base fluid.

Item	**Properties**	**Unit**	**Content**
1	Volume	ml	350
2	Mud weight	gr/cm3	1.121
3	O/W Ratio	-	80/20
6	PV	mpa.s	19
7	YP	pa	5.27
8	Gel 10"	pa	0.96
9	Gel 10'	pa	1.44
10	AV	mpa.s	24.5
12	HPHT Fluid loss	ml/30min	3.4

Table 3. Formulation of the OBDF containing nanofluid.

Item	**Product**	**Basic Fluid (Sample1)**	**Sample2**	**Sample3**	**Sample4**	**Sample5**	**Sample6**	**Sample7**
1	Calcium Chloride brine [ml]	81	81	81	81	81	81	81
2	Gas oil [ml]	227	227	227	227	227	227	227
3	P.Emulsifier [ml]	8.3	8.3	8.3	8.3	8.3	8.3	8.3
4	S.Emulsifier [ml]	6.6	6.6	6.6	6.6	6.6	6.6	6.6
5	FLC [gr]	12	12	12	12	12	12	12
6	Lime [gr]	10	10	10	10	10	10	10
7	Oil-based Viscosifier [gr]	1	1	1	1	1	1	1
8	Limestone [gr]	63	63	63	63	63	63	63
9	Nanofluid [V% of the brine]	0	1	3	5	7	9	11

megapascal) and room temperature (25 °C) using a rotary viscometer. According to the American Petroleum Institute (API) standards, the following equations were used to calculate plastic viscosity (PV), YP and AV.

$$PV = \theta_{600} - \theta_{300} \quad (1)$$

$$YP = \theta_{300} - PV \quad (2)$$

$$AV = \frac{\theta 600}{2} \quad (3)$$

where, θ300 and θ600 were measured at 300 and 600 rpm by reading the viscometer dial plate, respectively. Before measuring the rheological properties, the OBDF samples were mixed for 10 minutes to achieve steady-state conditions.

Filtration test

HPHT filtration experiments were performed using a HPHT filter press device with a CO_2 pressure regulator and a standard filter paper. This test was carried out at pressure of 500 psi and temperature of 300 °C with the standard method recommended by the API for testing OBDFs. The volume of filtrate was measured using a graduated cylinder.

RESULTS AND DISCUSSION

Characterization

The XRD pattern of CuO NPs is shown in Fig. 1. The peaks at angles 2θ= 32.39, 35.40, 38.62, 48.65, 53.30, 58.10, 61.44, 65.68, 66.17, 67.93, 72.30 and 74.99° indicate that the particles contain CuO crystals. The XRD spectrum confirms the formation of a single phase in which all the peaks can be indexed to the crystal planes of the CuO monoclinic structure (confirmed by the JCPDS card No. 45-0937) with the average crystallite size of 34 nm (Debye–Scherrer formula). No characteristic peaks of other impurities are observed that indicating the formation of a pure phase of CuO.

The FT-IR spectrum of nano-CuO was scanned in the region of 400 to 4000 cm^{-1}. As shown in Fig. 2, the peaks at 511 and 599 cm^{-1} belong to stretching vibrations of Cu-O band [33] confirming the formation of highly pure CuO NPs. The absorption bands at 1638 cm^{-1} was bending mode of vibration of water molecule [33]. Also the absorption peaks in the range of 3400–3700 cm^{-1} were due to the presence of O-H stretching vibrations [34] that represents water as moisture in the sample. The IR bands observed in the regions mentioned above confirmed the formation of CuO in the nanophase, which has also been reported by the several researchers [35].

The morphology characterization of the CuO NPs was investigated by SEM images as shown in Fig. 3 (a, b). The SEM images of CuO NPs showed that the surfaces of these microspheres (dandelion-like morphology) were composed of nanorods with an average diameter of about 40~55 nm, and almost all of them possess the same morphology.

Effect of pH on zeta potential of CuO NPs

Zeta potential value of the colloidal particles shows the stability or instability of the colloidal

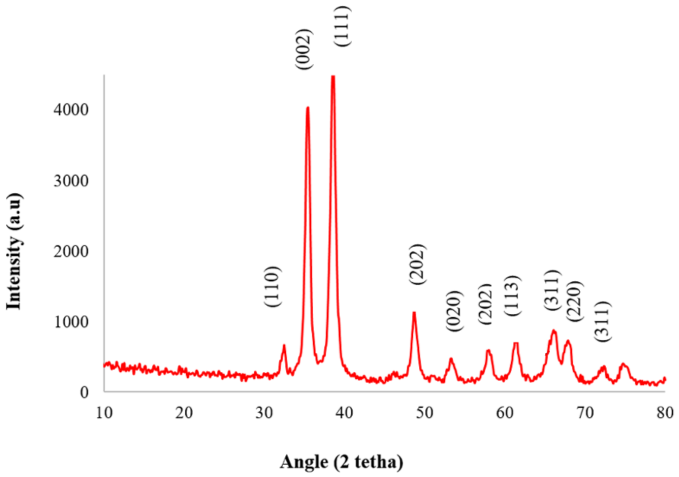

Fig. 1. XRD pattern of CuO NPs.

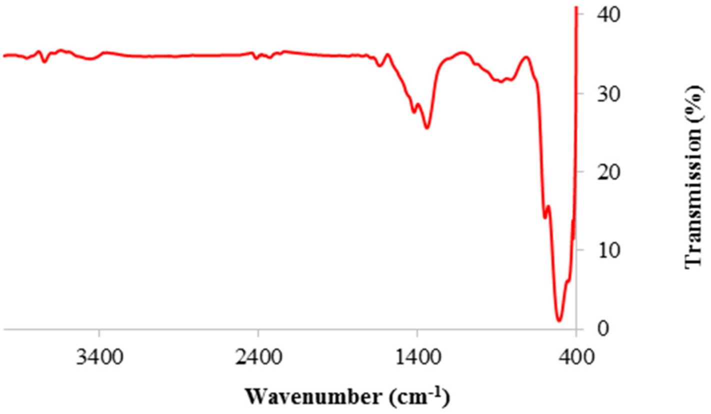

Fig. 2. FTIR of CuO NPs.

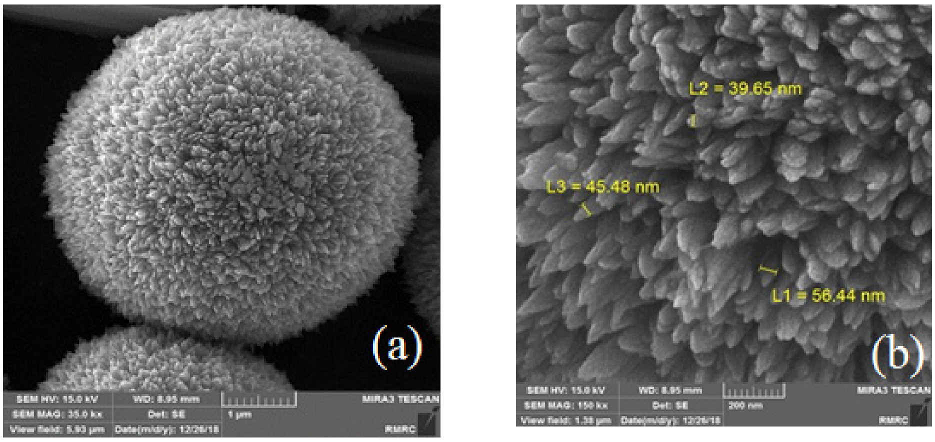

Fig. 3. SEM images of CuO NPs with dandelion morphology (a) magnitude of 1 μm (b) magnitude of 200 nm.

Table 4. Zeta potential values for CuO NPs at different pHs.

NP	pH=3	pH=6	pH=7	pH=8	pH=13
CuO	-1.79 [mv]	-17.9 [mv]	-25.6 [mv]	-29.8 [mv]	-19.7 [mv]

system. Indeed, if the zeta potential value of the colloidal particles is more than + 30 or less than -30 millivolt (mv), the colloidal system is stable [36]. The zeta potential is a function of the surface electrical charge density. Since the surface electrical charge is a function of pH, the value of zeta potential will be a function of pH. The effect of pH on the zeta potential of CuO NPs is shown in Table 4. pH adjustment was performed with 1 M NaOH or 1 M HCl. Considering Table 4, a pH of 8 was chosen for the working condition to ensure good and stable dispersion of CuO NPs.

Rheological properties of OBDF samples with and without CuO NPs

Fig. 4 shows the rheological characteristics of the OBDF with and without CuO NPs as an enhancing additive. Nanofluids containing 0.22% wt CuO NPs were added to OBDF ranging from 0-11% (V/V). The results revealed that the maximum value of PV was 26 millipascal-second (mpa.s) for the OBDF is containing 7% to 9% the NF. By an increase in the volume percentage of the NF to 11%, the PV value decreased to 25 mpa.s. In general, the NF additives may dilute or thicken

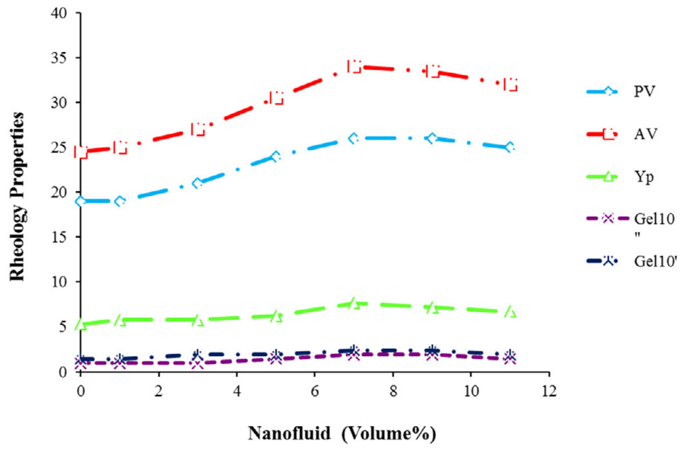

Fig. 4. The rheological characteristiccs of the OBDFs.

the drilling fluids which related to the type of NPs, type of the drilling fluid (water base or oil base) and the concentration of NPs in the NF.

According to Salih and Bilgesu, the PV value of the drilling fluid may be decreased by the addition of NPs. He showed that the addition of 0.5 to 1.5 pound per barrel (ppb) nanosilica to the drilling fluid decreased the PV from 28 to 24 centipoise (cp), respectively [6]. However, Esfandyaribayat et al. showed that the addition of CuO and Al_2O_3 NPs to the water-based drilling fluid increases the amount of PV. In contrast, by adding SiO_2 and increasing its concentration, the PV value remained constant. Furthermore, they confirmed that the addition of TiO_2 to the drilling fluid decreases the amount of PV [37]. The rheological and filtration properties of water-based drilling fluid were also investigated by Perween et al. with addition of zinc titanate NPs to drilling fluids. They showed that the addition of zinc titanate NPs increased the PV value [38]. Ghanbari et al. [39] and Anoop *et al.* [40] also showed that addition of 0.5% and 2% silica NPs to drilling fluid increased the PV values, respectively. Considering the mentioned results, it was confirmed that the addition of NPs to the drilling fluid caused changes in the PV values with different trends. Noticeably, it should be considered that the PV values should be within the recommended range when the NF containing NPs are used as an enhancing additive.

The YP is refers to the cuttings carrying capacity of the drilling fluid [41, 42]. It is defined as the minimum force to change the drilling fluid gel condition to flowing state. The YP of the drilling fluid must be within the recommended range. Increasing the YP value above the allowable range leads to flocculation, an increase in the surge and swap pressure and the unnecessary loss of pump pressure. Also, decreasing the YP value below the allowable range causes the barite sag [6]. Referring to Fig. 4, an increase in concentration of the NF up to 7% resulted to increase in YP from 5.2 pascal (pa) (for base fluid) to 7.1 pa. As the concentration of the NF increased to 11%, the YP decreased to 6.7 pa. The increase of the YP after addition of NP may be contributed to higher attractive force achieved due to higher surface area of NP in drilling fluid [43].

Fig. 4 also depicts the trend of AV characteristic of the drilling fluid. The AV values increased from 24.5 to 34 mpa.s for OBDF containing 7% V/V NF. Then, as the concentration of NF increased to 11%, the AV value decreased to 32 mpa.s.

The GS refers to the ability of the drilling fluid to improve and maintain the gel structure when the drilling operation is stopped. This is an important property of the drilling fluid that must be within the recommended range to maintain the unnecessary circulation pressure required to restart the drilling operation [44]. Excessive or low GS may cause some problems such as difficulty of running of logging tools or formation of a too fragile gel [45]. Fig. 4 represents the variation of the initial (10 second) and final (10 minute) GS values of the OBDF versus the NF concentration. The GS values of the OBDF for the both tests (10 s and 10 min) have nearly a similar trend.

The value of the GS improved with an

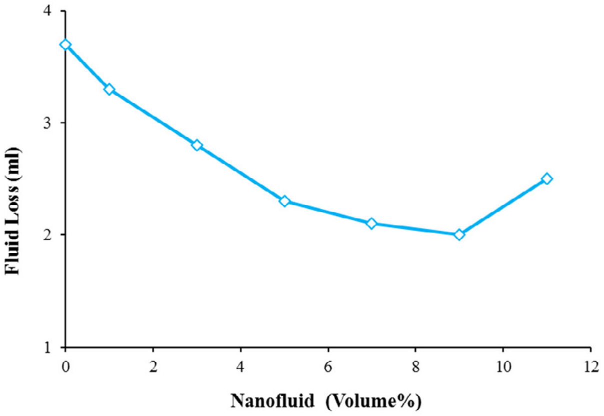

Fig. 5. The fluid loss of the OBDFs.

increase in the NF concentration representing an enhancement in the suspension capacity of the drilling fluid. The maximum values of initial and 10-minute GS were 1.9 and 2.3 pa for the both OBDFs containing 7 and 9% (V/V) NFs, respectively. In overall, the results proposed that the appropriate values of rheological properties belonged to the OBDF containing 7% (V/V) NF.

Filtration Properties under HPHT Conditions

Fig. 5 displays the results of the HPHT filtration tests conducted on the base fluid and the OBDF samples containing NF amounts of 1 to 11% (V/V). From Fig. 5, the fluid loss decreased by the addition of NF. The results showed that the minimum fluid loss value of 2 mL attained for the OBDF containing 9% (V/V) NF.

CONCLUSIONS

In the present research, the effects of adding CuO NPs with dandelion morphology to the OBDF were studied by measuring the rheological properties and HPHT fluid loss. Adding long time stable NF containing CuO NP to conventional OBDFs resulted to a low cost and high-performance OBDF with minimized formation damages. The results showed that the OBDF containing 7% (V/V) NF offered the best performance to improve the YP, AV, as well as the initial and 10-minute GSs. In addition, the OBDF containing 9% (V/V) NF had the minimum HPHT fluid loss of 2 mL. This study confirmed that using the synthesized CuO NPs was capable of assisting the drilling fluid in enhancement of its rheological properties and filtration characteristics. Furthermore, with addition of CuO NPs to OBDF formulation, the low volume of OBDF can be consumed; so, the released pollution to the environment will be reduced. In conclusion, a new path was proposed for the development of sustainable NF for the production of high-performance OBDF with HTHP applications.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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