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Colloidal Carbon Beads as Lubricant Additives in Water Based Drilling Fluid

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Abstract: *The friction related to bit wear, torque and drag greatly hinder the efficiency for a drilling operation. Colloidal carbon beads, synthesized by a facile solvothermal method, are investigated as an additive for improving the tribological performance. The application and tribological behavior of carbon beads as lubricant additives in water-based drilling fluid are estimated following a standard API/Baroid lubricity tester., the lubricity coefficient of composites of bentonite and carbon beads is lower even at a concentration of 0.2% (wt) compare to the concentration of 3% (wt) graphite powder. The coefficients of friction of carbon beads at different concentrations in water under 1.0 N normal load show significant improvement in its lubricity, reduced about 48% at the concentration of 0.8% (wt) compared to pure water. The rolling mechanism similar to nano or micro scale ball bearings of carbon beads can be pro-posed to account for the lower friction and wear performance.*

Keywords: *Carbon beads, Lubricant, Drilling fluid*

1. Introduction

Utilization of drilling fluids, a vital part and the lifeblood of a drilling operation, has never been more important than it is today in light of the deep or horizontal wells drilling for exploration of fossil fuels [1-3]. One of the main barriers of deep and horizontal well drilling operations is the high friction related to bit wear, torque between and drag, which usually causes drill pipe fracture and sticking accident[4, 5]. The performance, for instance rheology, fluid loss into the formation and lubricity, of drilling fluid highly relies on the chemical additives used in the system [6-9]. Drilling with oil-based drilling fluids offer less torque and drag, while for water-based drilling fluids, although environmentally safe and less costly compared to oil-based and synthetic-based fluids, high friction between the drill strings and wellbore is the vital issue to be concerned [10-15]. Although the current lubricant additives applied in water-based drilling fluids, in form of liquid or solid (e.g., saponated oil, emulsion and graphite etc.), exhibit good tribological behavior [16, 17], while concerns not only the sensitivity and stability on high temperature of liquid lubricants, but also poor dispensability in water of graphite powder, the performance of these lubricants in water based drilling fluid are greatly affected. Up to now, carbon-based lubricants (e.g., graphite powder, carbon nanotubes and nano-diamond etc.), with chemical inertia and excellent lubricity, have been widely applied in automotive, aerospace and industrial in solving routine lubrication problems other fields [18-24]. Furthermore, during the past decades, given the rapid advances in the synthesis of graphene in recent years, several preceding studies have been demonstrated graphene and graphene oxide sheets as additives in water-based lubricants with great enthusiasm [17, 25-28]. The sheets are believed to adsorb on the boundary surfaces, and act as protective coatings to reduce the friction and wear.

It is well known that the low-friction characteristics of solid lubricants are attributed to the slide between the layered structures on the molecular level; when the slip friction replaced by the rolling one with spherical particles, reducing mechanical loss, raising the efficiency, and promising extended application could be achieved. Consequently, inspired by ball bearings, spherical nano- and micro particles, become the promising candidate lubricants additives. Recently, nano- and micro sized particles as lubricant additives have gained considerable attention in light of their tribological properties and friction reduction mechanisms. Various inorganic nanoparticles have been studied as lubricant additives in thin film lubrication. These studies have reported reduced friction and wear in tribo-surfaces with the use of nanoparticle lubricant additives [5, 29-31]. For instance, Battez et al. demonstrated the reductions in friction and wear under extreme pressure of the CuO, ZnO and ZrO_2 nanoparticles mixed with base oil [32]. In addition, Vilt et al. investigated the tribological properties of silica micro spheres and showed direct evidence of the rolling mechanism of spherical particles [33]. Recently, Sadeghi et al. Prepared carbon micro spheres via an ultrasonic assisted method, and substantial reduction in friction and wear

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of carbon spheres and lubricating oils mixture were well discussed [34].

However, the application of nano or micro sized particles as lubricant additives in water based fluids is relative rare. Pesika et al. reported surfactant-functionalized carbon spheres as a new class of aqueous-based lubricants, which exhibited low friction coefficient and surface wear even at high loads and high contact pressures [35].

Herein, we report the preparation of sub-micro colloidal carbon beads with diameters of 300 to 500 nm via a modified established facile solvothermal method [36], and employ as an effective lubricant additive for water-based drilling fluid. Colloidal carbon suspensions exhibit high stability in aqueous system which attribute to the abundant hydrophilic functional groups on the surface (e.g. hydroxyl group, carboxyl group, aldehyde group et al..). The lubricity test, following the API standard, of CBs in base water drilling fluid show greatly reduction in lubricity coefficient (11.6-46.5%) with concentration range from 0.1% to 0.5 wt %. The coefficient of friction for carbon beads suspension in the concentration of 0.8 wt% is significant reduced around 48% compared with pure water.

2. Experimental methods

2.1. Materials

All chemicals used in the study were of A.R. grade and were purchased from Sino pharm Chemical Reagent Co. Ltd, China. The bentonite clay was obtained from Huawei bentonite Co., China. Graphite powder was purchased from Tianjin Guangfu Chemical Agent Co.

2.2. Preparation of colloidal carbon beads

The carbon beads (CBs) were prepared by using a modified previous reported method. Typically,15 g glucose was dissolved in 280 ml water and 20 ml ethanol, then the solution was transferred to a 450 mL Teflon-sealed autoclave and maintained at 180 ^oC for 6 hours. The resulting puce products were isolated by centrifugation, washed and purified by centrifugation and re-dispersion in deionized water and ethanol. Finally, the powder was dried at ambient temperature for further characterization and evaluation.

2.3. Lubricating property of CBs as additive in water based drilling fluid

The lubricity coefficient of the CBs in the base drilling fluid was determined using an OFITE, EP-Lubricity Tester. The bentonite solution was prepared by dispersed 40g as-received bentonite powder into 1L distilled water at a speed of 8,000 rpm stirring for 20min. Then the dispersions were pre-hydrated at room temperature for 24 h. To evaluate the lubricity of CBs in water based bentonite drilling fluid, the CBs were dispersed into bentonite aqueous suspension at different concentration by ultra-sonication. For the standard lubricity coefficient test,150 inch-pounds of force is applied between two test steel sur-faces, a block, and a ring rotating at 60 RPM, followed by the reading of the torque exerted by the fluid. With the torque obtained in a test performed with water, the correction factor (Cf) can be calculated according to the equation (1). Then, the suspension of bentonite and CBs was transferred to the equipment's container, carry out the above procedure to get the Reading fluid, then the lubricity coefficient (Lc) and lubricity coefficient reduced rate (ΔLc) can be attained according to the equation (2) and (3).

$$C_f = \frac{34.0}{Reading_{water}} \quad (1)$$

$$L_c = \frac{C_f * Reading_{fluid}}{100} \quad (2)$$

$$\Delta L_c = \frac{L_{c0} - L_{cl}}{L_{c0}} * 100\% \quad (3)$$

(L_{cl}: with lubricant, L_{c0}: without lubricant)

2.4. Tribological Study

To investigate the lubricity, uniform CBs were ultrasonically dispersed in water at different 0.1, 0.2, 0.4, and 0.8wt %concentrations while pure water as reference. The tribological property was performed at room temperature using a pin-on-disk apparatus (CSM Tribometer) which include the rotational motion of the disk with a stationary pin. A 4.2mmdiameter stainless steel ball was used as the stationary pin specimen while steel disk with surface roughness of 100 nm is rotating with a speed con-trolled by a DC servo motor. In a typical test, the pin-on-disk was immersed in10 ml CBs aqueous solution, the steel ball slid on the steel disk for 1000 shear cycles (the sliding diameter is 1.0 cm) under a constant load of 1.0 N, at a sliding speed of 5cm/s. All the tests were repeated twice for each concentration.

2.5. Characterization

The morphologies of the samples were examined by using a HitachiS-4800 field-emission scanning electron microscope (SEM). The FTIR spectra of sample were recorded in KBr pressed pellets on a Bruker IFS 66V/S FTIR spectrometer (Germany) in the mid-IR region of 400–4000 cm^{-1}.TG analysis was performed using a Netzsch STA 449C thermal ana-lyzer (Germany) at heating rates of 10^oC/min at temperature ranging from ambient temperature to 850^oC under N_2.

3. Results and discussions

Scanning electron microscopy (SEM) images of the carbon beads are presented in Figure 1. The results reveal that, in the presence of ethanol, the carbon spheres possess an average diameter of 300−500 nm, and individual spheres are well identified from each other without any agglomeration. Moreover, high-resolution SEM shows the smooth surface of the CBs, and the insert photo of CBs aqueous solution further indicates the high dispensability of CBs in water. Differentiate from the hydrophobic feature of graphite powder, high-dispersed in water greatly availed its application in aqueous system.

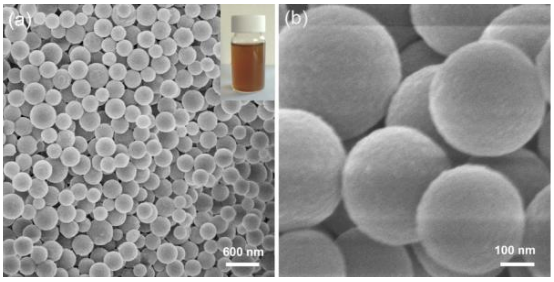

***Figure 1**. SEM images of colloidal carbon beads. Insert is carbon beads aqueous solution*

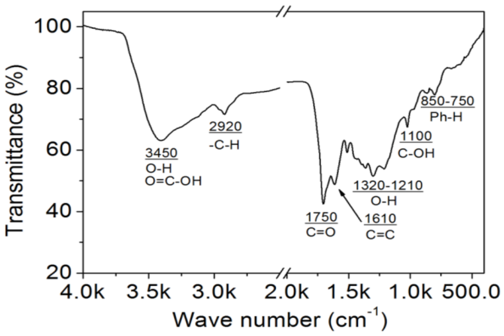

***Figure 2**. FT-IR spectra of as-prepared colloidal carbon beads*

The FT-IR spectrum is shown in Figure 2. Various hydrophilic functional groups on the CBs were observable. The characteristic adsorption bands belong to C=C stretching of aromatic (1610 cm^{-1}), carbonyl groups (1750 cm^{-1}), C–H stretching (2920 cm^{-1}), respectively. The red shift from 1725 cm^{-1} to 1650 cm^{-1}, which are attributed to the conjugation of aromatic ring or a, b-unsaturated double bonds, suggested that aromatization occurred. The band at 850–750 cm^{-1} corresponding to aromatic C–H out-of-plane bending vibrations also indicates the aromatization of glucose during the solvothermal treatment. The bands at 1100 $cm^{-1}$and 3450 cm^{-1} can be attributed to the C–OH bending vibrations and O–H (hydroxyl or carboxyl) stretching vibration, which indicating that large numbers of –OH groups were on the CBs surface. Overall, the FT-IR results indicate that aromatization occurred during solvothermal treatment, and that –OH and C=O are the major oxygen-containing functional groups on the CBs surfaces, which further verified the highly dispensability in water of CBs.

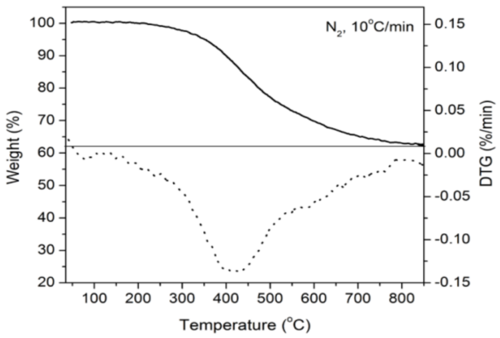

***Figure 3**. TG and DTG curves of colloidal carbon beads in nitrogen atmosphere*

In order to investigate the thermal behaviors of the CBs obtained from the solvothermal reaction, TG analysis was performed range from room temperature to 850 ^oC under N_2. From Fig. 3 it can be seen that the mass loss begins at around 200 ^oC, and the primary weight loss take place in the temperature range of 240–420 °C which could be attributed to the dehydration and densification of the CBs. Up to 450 ^oC, the weight loss is around 12%, indicating the oxygen and hydrogen content of colloidal CBs and showing the enough stability for application in high temperature drilling fluid.

To estimate the lubricity features of the CBs in water-based drilling fluid, bentonite suspension with concentration of 1% and 3% was pre-hydrated and used as base mud. Commercial graphite was used as the control sample; the concentration of 1%, 2% and 3% was added into the pre-hydrated bentonite suspension and stirred at speed of 8000rpm. Bentonite/CBs suspensions were also prepared following the above procedure. In Table 1 are presented the Lc and ΔLc of bentonite suspension, bentonite/graphite, and bentonite/CBs at various concentrations. The Lc of base mud is influenced by the content of clay and lubricant additives. Lubricity coefficient of commercial graphite powder is clearly decreased with the addition of graphite powder concentration range from 1% to 3%, and the lubricity coefficient reduced rate can be 37.2%, but it showed that little improvement in torque reduction and leveling off beyond a concentration of 2%. Notably, addition of CBs to base mud showed significant improvement in the lubricity. Even in the concentration of 0.2% (wt), the lubricity coefficient reduced rate can be 46.5%, along with the lubricity coefficient decreased from 0.43 to 0.23, implying the high lubricity at a low addition. In the concentration of 3% (wt) of bentonite suspension, the lubricity coefficient reduction of 47.8% can be obtained by addition of 5% (wt) carbon beads, further validating the effective performance of CBs in water-based mud.

To validate the possible ball bearing effect mechanism for CBs lubricity, SEM images of bentonite/graphite and bentonite/CBs are present in Fig.5. For bentonite/graphite mixture, it is clearly seen that the graphite particles (white dotted circle) and bentonite (black dotted circle) are isolated. On the other side, the CBs were tightly embedded in the interlayer of hydrated bentonite, which may be attributing to the abundant surface hydrophilic groups. The illustrations on the right side show the possible condition of the two kinds of compositions. For lubricity performance test, the CBs were supposed inserting into the hydrated bentonite layer and act as nano ball bearing, resulting in the enhanced lubricity property even at lower concentration.

Table 1. *The L_c and ΔL_c of base mud with various concentration of lubricant additives*

Clay	Lubricant (wt%)	L_c	ΔL_c (%)
Bentonite 1%		0.43	
	Graphite powder		
	1	0.37	13.9
	2	0.28	34.9
	3	0.27	37.2
	Carbon beads		
	0.05%	0.38	11.6
	0.1	0.32	25.6
	0.2	0.23	46.5
	0.5	0.22	48.8
Bentonite 3%		0.46	
	Carbon beads		
	0.1	0.41	10.9
	0.2	0.30	34.8
	0.5	0.24	47.8

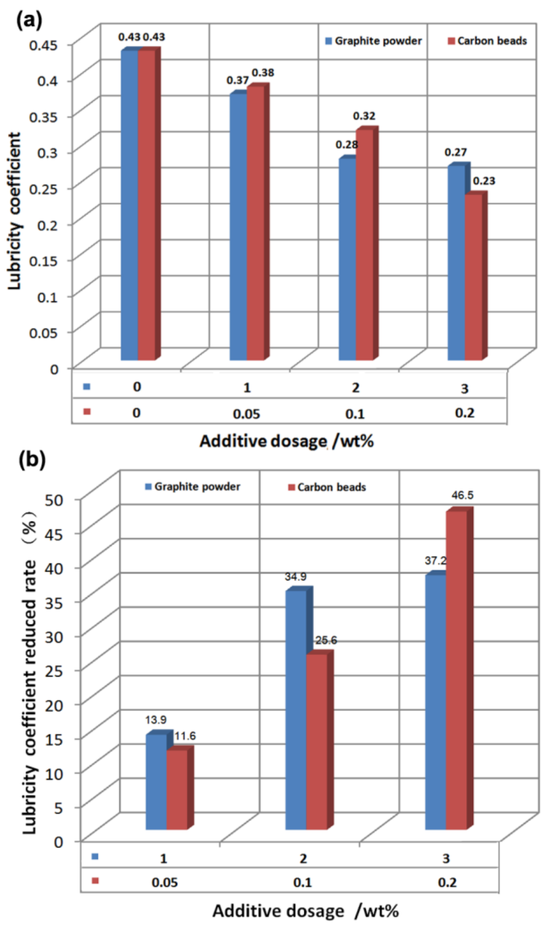

Figure 4. *Lubricity coefficient (a) and lubricity coefficient reduced rate (b) of graphite powder and carbon beads in bentonite suspension (1 wt %) with different concentrations*

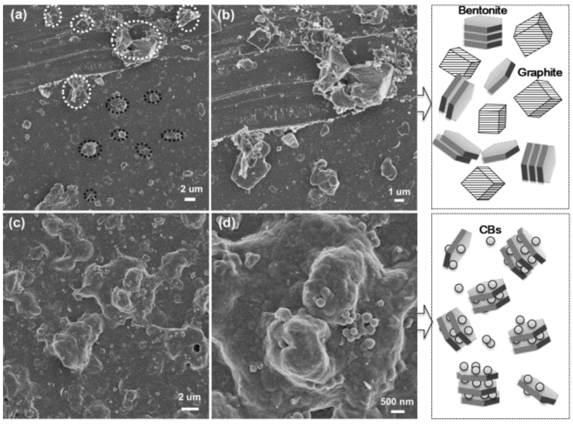

Figure 5. *SEM images of bentonite/graphite and bentonite/CBs suspension dropped on silicon wafer and dried at room temperature*

For further verification of lubricity of CBs in aqueous solution, the tribological performance was investigated using pin-on-disk (POD) tribometer. The typical measurement consisted of applying an initial pre-load of 1.0 N on the steel probe against a stainless steel disk with surface roughness around 100 nm. The sliding speed was set as 5 cm/s and the rotating diameter was 1cm. The effect of concentrations of CBs in water on the tribological performance ranging from 0.1% (wt) to 0.8% (wt) was studied. As shown in Figure 6, it can be seen that the CBs additives greatly reduced the coefficient of friction compared to water alone. The aqueous solution containing 0.8 % (wt) carbon beads exhibited approximately 50% reduced coefficient of friction.

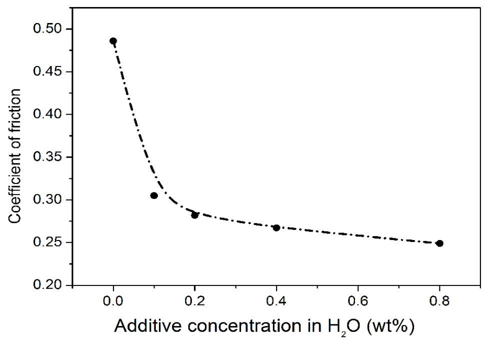

Figure 6. *Coefficient of friction for various concentrations of carbon beads in water under 1.0 N normal load (Sliding speed : 5 cm/s, rotating diameter: 1 cm)*

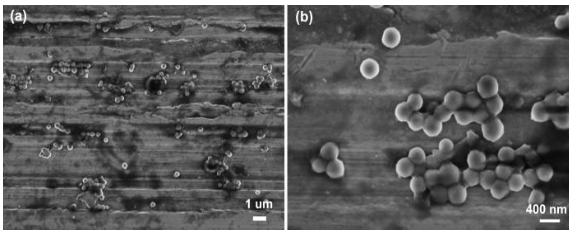

Figure 7. *SEM images of the wear scar on steel disk after sliding under 1.0 N normal load for 40 min*

The mechanical stability is another crucial issue of the solid additives since the lubricating surfaces generally are in contact with each other despite the fact that a fluid is existent in the boundary lubrication regime, which means the gap between surfaces in contact can be filled with solid lubricant additives when two surfaces come into contact because of surface roughness. And thus the prolonged use will drastically damage the lubricant between the gap due to the applied force. SEM images present in Fig. 7 are collected after 40 min test under 1.0 N applied load on the pin-on-disk apparatus. Obviously, CBs maintained their perfect spherical morphology, further implying that their rolling motion during tribological measurements. And this superior mechanical stability of micrometer carbon spheres makes them an excellent candidate as solid lubricant additive in aqueous system.

4. Conclusions

Colloidal carbon beads are demonstrated as lubricant additives in water based drilling fluid. A facile solvothermal strategy was performed to prepared the water dispersed carbon beads with a diameter ranging from 300 to 500 nm. The thermal and mechanical stability of the CBs makes them be regarded as the promising solid lubricant applied in water-based drilling fluid. The CBs are shown to have efficient improvement in the lubricity for the bentonite base mud at lower concentration. Moreover, pin-on-disk tribological tests validated a remarkable reduction in friction and wear (~48%) by adding 0.8 wt % of carbon beads in water. The results presented herein can be explained by the possibly rolling motion on the nanoscale where CBs act as ball bearings to lower friction and wear in the boundary regimes. It is anticipated that this study can give an impetus to pursue to other high efficient solid lubricant additives for water-based drilling applications.

5. Acknowledgments

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