Organic-Sulfonate Functionalized Graphene as a High Temperature Lubricant for Efficient Antifriction and Antiwear in Water-Based Drilling Fluid

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Abstract

The lubricity of drilling fluid resistant to high-temperature over 200°C is still one of the technological breakthroughs. In this study, the graphene modified with sodium dodecylbenzene sulfonate (SDBS) was selected as a resistant to high-temperature lubricant. Our results show that the drilling fluids have high stability after aging at 240°C with the assistance of the SDBS/graphene. Excitingly, the tribological performance test results revealed that the SDBS/graphene exert excellent anti-friction and anti-wear properties. Compared with the base slurry, the friction coefficient and wear rate of the SDBS/graphene slurry are reduced by 76% and 59%, respectively. The deposited film composed of graphene, Al₂O₃, SiO₂, Fe₂O₃, FeSO₄ actualized the protection of the sliding contact zone, proving that the sulfonate group on the SDBS/graphene contributed to prompt the deposition of the graphene and bentonite and then enhanced tribological properties of the drilling fluids. Overall, the graphene modified with SDBS is expected to solve the difficulty to form effective deposited film and poor lubricity of the drilling fluid under high-temperature.

1 Introduction

During the drilling and extraction process of ultra-deep wells, the contact between the drilling pipe and casing made the casing severe worn[1, 2] and increased the power consumption[3]. Moreover, the lubrication properties of drilling fluid under the temperature of ultra-deep wells bottom hole above 200 °C would become invalid and may further cause collapse[4–6]. Hence, it is in urgently need to reduce friction and wear during oil and gas drilling operations under high temperature[7, 8]. Polymer materials are being extensively utilized as additives for enhancing lubrication performance at high-temperature[9–11]. Polycarpou et al. reported the aromatic thermosetting copolyester coating could increase the temperature resistance to 175 °C[12]. However, the lubrication effects of the polymer under the higher temperature above 200 °C were sharply dropped, due to the polymer degraded and cross-linked. Hence, developing a facile approach for boosting the lubrication activity of drilling fluids in high temperature over 200 °C is urgently needed.

In recent years, carbon based lubricants have been widely used in water-based drilling fluids for their excellent properties such as high temperature resistance, non fluorescence and good lubrication performance. Among them, graphene has excellent mechanical[13] and easy shear capability between stacked lamellar structure[14, 15]. However, graphene cannot be directly used in the water-based drilling fluids for its intrinsically hydrophobicity. Graphene oxide[16] and reduced graphene oxide[17] have attracted enormous research interest in lubrication additives application. However, the oxygen-containing functional groups would hinder the sliding of graphene layers. There is still a lack of effective lubrication activity in water-based drilling fluids. We prepared SDBS/graphene by a shear exfoliation assisted with supercritical carbon oxide (SC-CO₂) fluid[18]. The yield of the prepared graphene was as high as 63.2% and the intensity ratio of D peak and G peak of the prepared graphene is 0.31, which is much less than 1.1-1.5 of graphene prepared by redox method[19]. The SDBS not only endowed graphene with hydrophilicity and effectively prevent graphene layers agglomeration by providing electrostatic repulsion
or steric hindrance[20], but the SDBS also had a certain anti-wear performance[21]. Therefore, the SDBS/graphene prepared by supercritical carbon dioxide would present potential resistance to high temperature lubrication performance in water-based drilling fluid.

Here, we developed a new strategy by introducing the SDBS/graphene into water-based drilling fluid to effectively antifriction and antiwear at room temperature of 25 °C and at increased temperature of 240 °C. The rich sulfonate groups on the SDBS/graphene surface made bentonite easy for physisorption and subsequent deposition on friction pairs. Importantly, the hydrophily nature enables the homogeneous distribution of graphene inside drilling fluids to produce a uniform robust deposition film. As expect, the prepared graphene presents excellent the tribological behaivors to inhibit friction and wear under aging at high temperature of 240 °C. Overall, our design holds great promise for high temperature resistant drilling fluid lubricant additives.

2 Methods

2.1 Material

Graphite powder (99.90%) and SDBS (88.00%) were purchased from Sinopharm Chemical Reagent Co., Ltd. Carbon dioxide (99.90%) was purchased from Shanghai High-tech Co., Ltd. Absolute ethanol (99.50%) was purchased from Changshu Yangyuan Chemical Co., Ltd. Sodium-bentonite was purchased from Hongfa bentonite factory (Xinyang, China). Sodium carboxymethyl cellulose and anhydrous sodium carbonate were provided by Tianjin Komi Chemical Reagent Co., Ltd. Deionized waters were used in all experiments.

2.2 Preparation of graphene dispersion

The SDBS/graphene powders were carried out according to the previous report[19]. The powders were dispersed into water with a concentration of 0.7 mg/mL. The graphene dispersion was kept standing 24 h to remove precipitate and then freeze-dried.

2.3 Preparation of the drilling fluid

The base slurry of water-based drilling fluid was consisted of 4 wt.% sodium bentonite (Na-Mt), 0.5 wt.% sodium carbonate and 0.1 wt.% sodium carboxymethyl cellulose. A certain mass of SDBS/graphene was added to the base slurry. The mixture was stirred at 8000 rpm for 20 min, and then standing for 24 h for the hydration of bentonite. Aging tests were conducted by hot rolling in a roller oven (Qingdao Senxin Electromechanical Equipment Co., Ltd.) at different temperatures for 16 h.

2.4 Evaluation of drilling fluid properties

The zeta potential of the drilling fluids were measured by a Zetasizer Nano (Malvern ZS90, UK). The lubrication coefficients of the drilling fluids were investigated using an Extreme Pressure (EP) lubrication instrument (Qingdao Senxin Electromechanical Equipment Co., Ltd.). The friction coefficients, wear rate and the extreme pressure experiments of the drilling fluids were evaluated using reciprocating mode by
the micro-friction testing machine (SRV-5, Germany Optimol) under a temperature of 25 °C, load of 50 N, frequency of 1 Hz and stroke of 1 mm for 0.5 h. The upper ball was GCr15 steel with an elastic modulus of 205 GPa and a Poisson ratio of 0.30. The disc was 304 stainless steel with Φ 24 × 7.9 mm and an elastic modulus of 194 GPa. The wear rate of the wear surface was obtained by using three-dimensional (3D) profile (Bruckner, USA). X-ray photoelectron spectrometer (XPS, Kratos, UK), Renishaw Raman microscope and Scanning electron microscope (SEM, Carl Zeiss, Germany) were used to characterize the chemical state and element surface distribution of typical elements on the wear surface.

3 Results And Discussion

3.1 The morphology and structure of graphene

As shown in Fig. 1a, the prepared graphene was wrinkled, thin and transparent. The G peak at ~ 1580 cm\(^{-1}\) and the 2D peak at ~ 2700 cm\(^{-1}\) can been clearly observed (Fig. 1b). The D peak appeared at 1350 cm\(^{-1}\) is weak, indicating that the disorder carbon degree of the prepared graphene was low. In Fig. 1c, the sodium bentonite exhibited a weight loss at approximately 65°C, which was attributed to the loss of free and interlayer water adsorbed in the interlayer of the Na-Mt[22]. However, no mass loss in the TGA curve of graphene before 340 °C, which demonstrates that sulfonic acid anion intercalated into the interlayer space and replaced the interlayer water molecules[23]. The weight loss between 340 °C to 370 °C was assigned to the decomposition of sulfonic acid groups. The borehole temperature during drilling was far below 300°C, so the modified graphene would not be thermally decomposed underground[24]. The zeta potential of the base slurry is -119.70 mV (Fig. 1d). After adding the prepared graphene, the zeta potential value of the drilling uid is -158.70 mV, which shows that the prepared graphene had high dispersion in water-based drilling fluids. It was attributed to the electrostatic repulsion between the sulfonic acid groups in the SDBS and the base slurry and then promoted the diffusion of cations in the clay particles into the water-based drilling fluid, the increased ionization degree of the clay made the tight combination of graphene and the clay.

3.2 The tribological properties of drilling fluids

Figure 2a shows the lubrication properties of the drilling fluids. With the increasing mass ratio of graphene, the lubrication coefficient of the drilling fluids dramatically decreased to 0.15 at 1%. Beyond 1%, the decreasing extent reduced and reached 0.14 at 2%, which suggests that a mass ratio of 1% was enough to achieve effective lubrication. The decreased lubrication coefficient was attributed to the formation of a tribo-chemical reaction film resulting from an increase mass of the obtained graphene. At a higher mass, more graphene sheets can combine with clay by electrostatic repulsion to form a protective film reducing direct contact in friction pair[25–27]. To future clarify the enhanced lubrication, the influence of SDBS with drilling fluid on lubrication coefficient was studied. The lubrication coefficient decreased from 0.49 to 0.30, which could be ascribed to the role of the SDBS in boundary lubrication. The influence of the aging temperature was studied when the mass ratio of graphene was fixed at 1%. As shown in Fig. 2b, the lubrication coefficient of the base slurry increased from 0.74 to 0.16. when the
temperature increased from 180 to 240 °C, whereas the lubrication coefficient of the drilling fluid with graphene first decreased to 0.11 at 230 °C and then remained unchanged after increasing to 240 °C. This result suggests that the aging temperature affects dramatically the lubrication of the drilling fluids. When the aging temperature was too high, the agglomeration of clay particles in base slurry was worse, resulting in the difficulty for clay to absorb on the surface of friction pair. While the clay combined with graphene sheets, the formation of the deposited film was not affected at the elevated temperature. The lubrication coefficient of the base slurry with 1 wt.% graphene was 0.11 and 0.16, respectively.

To explore the influence of the graphene sheets and SDBS on the antifriction properties of the drilling fluids, the base slurry and the base slurry with graphene, SDBS were systemically investigated by an SRV-5 testing machine. Fig. 2c shows the friction coefficient of pure base slurry was about 0.80, and the friction coefficient and the running time quickly decreased with the increasing graphene content. Beyond 1%, the decreasing extent reduced and reached 0.09 at 2%. After aged at 240 °C, the running time and friction coefficient of the base slurry with 1 wt.% graphene were reduced from 400 s to 20 s, 0.85 to 0.20, respectively. It indicated that graphene could quickly form a tribo-film on the surface of the friction pair under high temperature.

The antifriction properties of the SDBS were measured in a water and base slurry. As presented in Fig. 2d, the friction coefficient of the SDBS in water was higher than that of the graphene in water and the base slurry with 1 wt.% graphene. The average friction coefficient of the SDBS in base slurry was 0.70, which was 4 times higher than that of the base slurry with 1 wt.% graphene (0.16). This suggested that the graphene has a more effective antifriction property. The filtrate loss of the drilling fluids at room temperature were recorded and as shown in Fig.S1, there was not too obvious change in the filtrate loss with the increasing graphene contents. When the aging temperature increased to 180~250 °C, the filtration loss of the drilling fluid was 3 times higher than that of the room temperature (Fig.S2).

To further probe the anti-wear properties of the drilling fluids, the three-dimensional profiler was used to observe the three-dimensional morphology and the wear rate of the wear scar. As shown in Fig. 3a, it can be seen that the wear scar of the base slurry was deeper and rougher than that of the graphene drilling fluids. The wear rate of the base slurry was 2.98×10^{-6} mm³/N·m. Compared to base slurry, the wear rate of drilling fluids with 0.5% graphene decreased by 49% (1.53×10^{-6} mm³/N·m), by 69% for 1% graphene (9.3×10^{-7} mm³/N·m) and by 63% for 2% graphene (1.09×10^{-6} mm³/N·m). This was attributed that excessive graphene sheets would aggregate with bentonite particles, which impeded the formation of a tribo-film and influenced the tribological performance. The wear rates of the water with 0.25% SDBS, drilling fluid with 0.25% SDBS, water with 1% graphene were 5.13×10^{-6} mm³/N·m, 3.54×10^{-6} mm³/N·m and 5.82×10^{-6} mm³/N·m, respectively. The results indicated the bentonite and graphene played an important role in the anti-wear performance of drilling fluid. After aging at 240 °C, the wear scar of the drilling fluid with 1% graphene was obviously shallower than that of the base slurry. The wear rate of the drilling fluid with 1% graphene (1.24×10^{-6} mm³/N·m) reduced by 59% compared with the base slurry
(3.06×10^{-6} \text{ mm}^3/\text{N}\cdot\text{m})$, this indicated that graphene could significantly improve the anti-wear of the drilling fluids under high temperature of 240 °C.

We used the SEM and EDS to analyze the morphology of the wear scar and the element composition of the tribofilms. As shown in Fig. 4, a large number of furrows and dents appeared on the wear scar surface of the base slurry. Only a small amount of iron, oxygen and silicon deposited on the surface, which indicating that abrasive and oxidative wear occurred during the friction process. Compared to the base slurry, the wear scar surfaces of graphene drilling fluids were enriched with black contents, especially, that of 1% graphene was densest. It could be seen that massive iron, oxygen, silicon, aluminum and carbon were deposited on the wear scar surface of graphene drilling fluids, which showed graphene could facilitate the silicon and aluminum to deposit and then form the tribo-film on the friction surface. This was mainly due to the electrostatic attraction between sulfonate ions and iron ions, which made graphene sheets easy to deposit and combine with the bentonites on the surface of the friction pair. As shown in Fig.S3, large furrows and grooves occurred and a small amount of iron and oxygen deposited on the surface of the wear scar of graphene and SDBS dispersion, SDBS drilling fluid. Compared with the drilling fluid with 1% graphene, severer wear debris and grooves produced on the surface of the wear scar of the base slurry after aging at 240 °C. From the element surface distribution on the wear scar surface, similar to the room temperature, a large amount of iron, oxygen, silicon, aluminum deposited on the wear scar surface of graphene drilling fluid contains, while the carbon deposition changed inapparent. It indicated that the friction mechanism of graphene drilling fluid at high temperature maybe be the same as that at room temperature.

In order to further explore the element composition and chemical state of the worn surface of steel ball at room and high temperature, the XPS spectra of the graphene drilling fluid was analyzed shown in Fig. 5 and Fig.S4. For the sample at room temperature (Fig.S4), in Fe2p, the peaks at 709.8 eV (2p3/2) and 719.3 eV (2p1/2) belonged to Fe; the peaks at 710.9 eV (2p3/2) and 724.4 eV (2p1/2) were corresponded to Fe$_2$O$_3$, 712.9 eV (2p3/2), 726.3 eV (2p1/2) peaks were corresponded to FeSO$_4$[28, 29]. In O1s, 530.1 eV, 531.2 eV, 531.8 eV, 532.4 eV, 532.9 eV, 532.5 eV were corresponded to Fe$_2$O$_3$, Al$_2$O$_3$, SiO$_2$, C-O, C=O, FeSO$_4$, respectively. In Si2p, the peak at 102.5 eV was corresponded to SiO$_2$. In Al2p, the peak at 74.5 eV was corresponded to Al$_2$O$_3$. For S2p, the peaks at 167.2 eV and 168.8 eV belonged to S-O or S=O, the peak at 170.1 eV was corresponded to FeSO$_4$. In C1s, 284.8 eV and 285.2 eV were sp$^2$ and sp$^3$ hybrid carbon, respectively. The peaks at 286.4 eV, 287.1 eV and 288.5 eV were corresponded to C-O, C=S, and C=O, respectively[30]. Comprehensive the XPS analysis showed that graphene had excellent friction reduction and anti-wear performance, because graphene was first deposited and helped bentonite deposited on the surface of the friction pair to form a tribo-film composed of Al$_2$O$_3$, SiO$_2$ and C. In addition, a tribo-chemical reaction film composed of Fe$_2$O$_3$ and FeSO$_4$ formed during the friction process. Thus, the formation of the chemical reaction and deposited film greatly improved the tribological properties of the drilling fluid. Similarly, it could be seen that the Fe$_2$O$_3$, FeSO$_4$, Al$_2$O$_3$, and SiO$_2$ peak positions on the wear scar surface of graphene drilling fluid after aging at 240 °C (Fig. 5). However, the peak intensity of sp$^3$
hybrid carbon after aging at 240 °C slightly increased compared with that of the room temperature. This maybe attributed with the enhanced degree of graphene defects at high temperature.

The Raman was used to characterize the degree of graphene defects during the friction experiment at room temperature and aging 240 °C. As shown in Fig. 6, the intensity ratios of D peak and G peak ($I_D/I_G$) of graphene drilling fluid at room temperature and aging 240 °C aging were 1:2 and 1.62:1, respectively. $I_{2D}/I_G$ of that two sample were 0.34:1 and 0.25:1, respectively. In Fig. 1b, the $I_D/I_G$ and $I_{2D}/I_G$ intensity ratios of the graphene sample were 1:5 and 1:2, respectively. The results showed that the degree of graphene defects increased with the increasing aging temperature, thus the anti-friction anti-wear performance at aging 240 °C was reduced. In addition, the peaks at 534 cm$^{-1}$, 670 cm$^{-1}$, and 1475 cm$^{-1}$ were the characteristic peaks of Fe$_2$O$_3$, the peak at 802 and 1173 were consistent with SiO$_2$ and Al$_2$O$_3$, respectively.

### 3.2.3 Tribological mechanism

According to the analysis of the wear scars of samples aged at different temperatures, we inferred the tribological mechanism of graphene as an additive for drilling fluid shown in Fig. 7. During the friction process, the negative charged sulfonic acid groups of the SDBS connected with graphene by a π-π bond, not only electrostatically attracted the positive charges on the edge of the bentonite, but also formed an ionic bond with the iron substrate, which promoted the deposition of the bentonite on the sliding contact interfaces. Under the action of frictional shear, a tribo-film composed of graphene, Fe$_2$O$_3$, FeSO$_4$, Al$_2$O$_3$ and SiO$_2$ was formed to protect friction and wear. At high temperature, the sp$^2$ carbon was changed to sp$^3$, the deposited tribo-film of graphene would become discontinuous and thus decreased anti-friction and anti-wear properties.

### 4 Conclusions

In the present work, graphene functionalized with SDBS was used as a lubricating additive in water-based drilling fluid. It disclosed that graphene exhibited an excellent dispersibility in drilling fluid. Graphene improved significantly the anti-friction and an-wear performance at room temperature and high temperature at 240 °C. It is proposed that the graphene additive could facilitate the deposition of bentonite, which dramatically enhanced the roustness of the tribofilm. Thus, the graphene was found to be a pontential lubricating additive with enhanced tribological performances in water-based drilling fluid.

### Declarations

#### Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.
Conflict of interest There is no conflict to declare.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at.

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Figures

Figure 1
<p><strong>(a)</strong> TEM image of graphene, <strong>(b)</strong> Raman spectra of graphene, <strong>(c)</strong> TGA patterns of the Na-Mt and graphene, <strong>(d)</strong> Zeta potential values of the samples</p>

Figure 2
<p><strong>(a)</strong> The influence of graphene concentration and SDBS on the lubricity of water-based drilling fluid, <strong>(b)</strong> Lubrication coefficient of graphene-based drilling fluid at different temperature, <strong>(c)</strong> Friction curve of different samples of drilling fluid, <strong>(d)</strong> Friction coefficient curve of graphene-based drilling fluid and base slurry aged at 240 °C. <strong>(e)</strong> Lubrication performance of mud with 1% graphene after aging for 16 hours at 240 °C.</p>

Figure 3
<p><strong>(a)</strong> Three-dimensional morphology images, <strong>(b)</strong> The diagrams of the wear rate of different samples.</p>
Figure 4

The SEM and typical element distribution diagram of tribofilms. The six groups are (1) Base slurry, (2) Base slurry+0.5% graphene, (3) Base slurry+1% graphene, (4) Base slurry+2% graphene, (5) Base slurry at 240°C, (6) Base slurry+1% graphene at 240°C respectively.

Figure 5

The XPS spectra of steel black wear scars of the drilling fluid with 1% graphene after aging at 240 °C.

Figure 6

The Raman spectrums of the wear scar of graphene drilling fluids after aging at (a) room temperature and (b) 240 °C.

Figure 7

The tribological mechanism diagram of graphene drilling fluid at friction process.

Supplementary Files

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