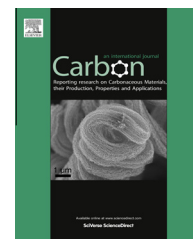


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Letter to the Editor

Tribological properties of monolayer graphene oxide sheets as water-based lubricant additives



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ABSTRACT

Application and tribological properties of graphene oxide (GO) monolayer sheets as additives in water-based lubricants were investigated. The lubricating fluids were applied to a sintered tungsten carbide ball and stainless steel flat plate. It was found that adding GO particles into water improved lubrication and provided a very low friction coefficient of approximately 0.05 with no obvious surface wear after 60,000 cycles of friction testing. GO adsorption occurred on the lubricated surfaces of both the ball and flat plate, suggesting GO sheets may behave as protective coatings.

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The use of water as a lubricating fluid has many advantages such as low cost, good environmental compatibility and high thermal conductivity. However water is a poor lubricant for steel and related materials [1], which are commonly used on the convectional contacting surfaces of machine elements. It is desirable to develop high-performance additives for water lubricants that may allow use with steel materials. Recently carbon nanomaterials have been studied as water lubricating additives, and have shown good tribological performance [2]. Hydrophilic treatments to carbon nanomaterials are necessary for their use as water lubricating additives, because the materials are innately hydrophobic. Such treatments increase the costs of using these additives. Graphene is a single atom thick, with a two-dimensional structure derived from a single layered sheet of graphite, and has recently attracted much attention for its unique properties, including lubricating potential [3,4]. Graphene oxide (GO) has been used as a precursor for large-scale production of graphene. GO has many carbon oxygen functional groups, which allow for dissolution in water. This means that

no hydrophilic treatments are needed. In this study, the tribological properties of single layer GO sheets dissolved in water between a sintered tungsten carbide (WC) ball and a stainless steel substrate were investigated.

Tribological tests were performed using a reciprocating sliding configuration with a sliding width of approximately 2.5 mm, a sliding frequency of 5 Hz, a load of 1.88 N, and 60,000 cycles. A Hertz contact pressure was estimated to be approximately 2.2 GPa under these frictional conditions. A SUS304 stainless steel flat plate and a WC ball with a diameter of 2 mm were cleaned by ultrasonication in acetone, ethanol, and finally in water for 5 min each. The volume of lubricant fluids used for the friction experiment was approximately 10 ml. To prevent evaporations for lubricant fluids, the ball and flat plate were covered with an acrylic box. In this study, lubricating fluids did not be added during frictional experiments. The lubricated surfaces of the ball and plate were investigated by scanning electron microscopy (SEM) and energy dispersive X-ray spectroscopy (EDX).

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GO used in this study was synthesized by modified Hummers' method. To induce oxidation, microwave irradiation was used during the oxidation process. The size of the GO sheets was distributed from 10 to 50 μm . Fig. 1 shows an atomic force microscopy (AFM) image and height profile of the GO sheets. The sample was prepared by dropping a 0.01 wt.% GO dispersion onto a freshly cleaved mica substrate followed by drying in air. The height of the GO sample was measured at around 1 nm. The distance between the graphene sheets in graphite is around 0.335 nm. As the surface of GO is composed of a large number of oxygen functional groups, the heights of GO measured by AFM are likely to be more than 0.335 nm (typically around 1 nm) [5]. From this was concluded that the GO sheets used in this study had a single monolayer structure.

The monolayer GO sheets were dissolved in purified water, at a concentration 1 wt.%. Purified water, an emulsified water-based liquid with an oil concentration of 1 wt.%, poly-alpha-olefin (PAO) were used as lubricating fluids for comparison. Fig. 2 shows the variations of friction coefficients when lubricated with these fluids as a function of friction cycles. Considering the friction speed of 0.025 m/s, boundary lubrications would be occurred in the frictional experiments. The friction coefficient of purified water was high at over 0.4. The water-based emulsion lubricant maintained a friction coefficient of approximately 0.12. At first a friction coefficient of the PAO was approximately 0.08, and decreased to around 0.06 at 10,000 cycles, and then increased to around 0.09. Three

measurements were conducted for the GO dispersion, with the same frictional condition. The friction coefficients of the GO dispersion were initially kind of high, and then decreased to be under 0.05. Friction cycles when the friction coefficients became approximately 0.05 were very distributed from 3000 cycles to 20,000 cycles in the three measurements of the GO dispersion.

Fig. 3 shows SEM images of the WC ball surfaces lubricated in purified water and the GO dispersion. The surfaces were washed by ultrasonic cleaning in ethanol after the friction tests. The wear on the WC ball surface lubricated with purified water could be clearly seen in Fig. 3(a). A circular wear scar of diameter $\sim 300 \mu\text{m}$ was evident. From the diameter of this scarring the wear depth of the WC ball surface was estimated to be approximately 6 μm . A high magnification image of the wear surface is shown in Fig. 3(b) indicating a loss of particles from the sintered WC surface and an increase in the surface roughness. The surface lubricated with the GO dispersion is shown in Fig. 3(c) and did not show any obvious signs of wear. However dark areas were observed on the surface (a high magnification image and carbon elemental mapping of the dark area are shown in Fig. 4(a) and (c), respectively). A high magnification image shown in Fig. 3(d) indicates that no WC particles were displaced from the surface lubricated with the GO dispersion. It was also noted that some areas of the surface in the high magnification images Fig. 3(d) appeared slightly darker with no apparent morphological changes.

Fig. 4(a) and (c) show typical SEM images of the ball and flat plate surfaces, respectively, lubricated in the GO dispersion after a friction test of 60,000 cycles. Fig. 4(b) and (d) show carbon element mappings measured by EDX, of the same areas in Fig. 4(a) and (c), respectively. The flat plate surface lubricated with purified water was scratched to a depth of approximately 6 μm (not shown). The SEM image of the WC ball surface lubricated with the GO dispersion shown in Fig. 4(a) showed slightly dark areas without any morphological changes, and also some much darker areas that

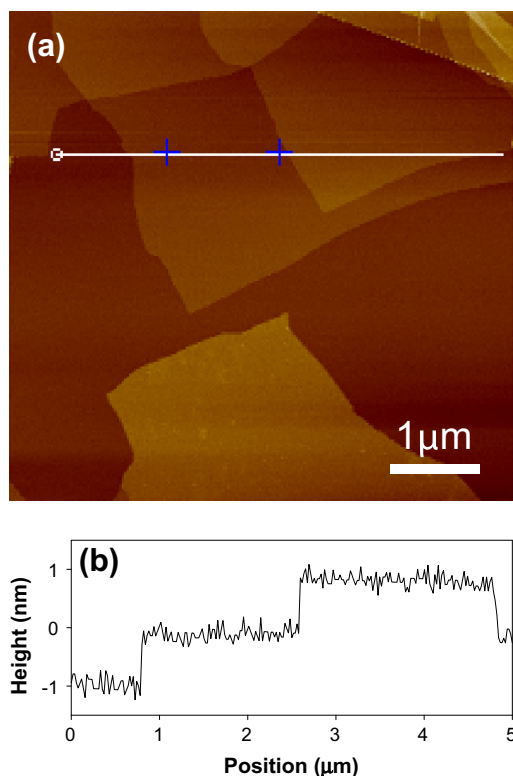


Fig. 1 – AFM image of GO sheets on a cleaved mica surface followed by drying in air. (a) 2D AFM image and (b) height profile corresponding to the line of the 2D AFM image. (A colour version of this figure can be viewed online.)

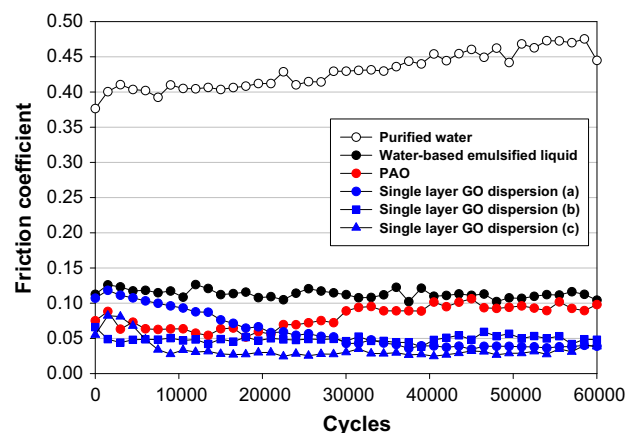


Fig. 2 – Variations of friction coefficients lubricated with purified water, the water-based emulsified liquid, PAO, and the GO dispersion, as a function of friction cycles, between the WC ball and stainless steel flat plate under the load of 1.88 N.

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