Role of nanoparticles in achieving macroscale superlubricity of graphene/nano-SiO₂ particle composites

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Abstract: Recent studies have reported that adding nanoparticles to graphene enables macroscale superlubricity to be achieved. This study focuses on the role of nanoparticles in achieving superlubricity. First, because graphene nanoscrolls can be formed with nanoparticles as seeds under shear force, the applied load (or shear force) is adjusted to manipulate the formation of graphene nanoscrolls and to reveal the relationship between graphene-nanoscroll formation and superlubricating performance. Second, the load-carrying role of spherical nano-SiO₂ particles during the friction process is verified by comparison with an elaborately designed fullerene that possesses a hollow-structured graphene nanoscroll. Results indicate that the incorporated nano-SiO₂ particles have two roles in promoting the formation of graphene nanoscrolls and exhibiting load-carrying capacity to support macroscale forces for achieving macroscale superlubricity. Finally, macroscale superlubricity (friction coefficient: 0.006–0.008) can be achieved under a properly tuned applied load (2.0 N) using a simple material system in which a graphene/nano-SiO₂ particle composite coating slides against a steel counterpart ball without a decorated diamond-like carbon film. The approach described in this study could be of significance in engineering.

Keywords: nano-SiO₂ particles; graphene; macroscale superlubricity; lubrication mechanism

1 Introduction

Superlubricity refers to a state in which the friction coefficient between two contacting surfaces is less than 0.01 or even close to zero and represents an ideal state sought after by many tribologists [1, 2]. Because of the potential to reduce friction and save energy, superlubricating materials are of great significance for energy saving and environmental protection [3, 4]. Currently, nanoscale and microscale superlubricity can be achieved by desirable incommensurate contact [5–11]. However, friction at the macroscale remains a complicated topic and encounters intricate factors that affect the lubricating performance of materials

[12–14]. In the past few decades, some tribologists have observed macroscale superlubricity in some systems and have proposed several related lubrication mechanisms [15–18] that could be helpful in understanding how to reduce friction and wear to a negligible level (even zero) in sophisticated industrial applications. Unfortunately, macroscale superlubricating mechanisms remain unclear at this time [19, 20], and further investigating the macroscale lubrication mechanisms of materials to better understand their superlubricating performance and to promote their application is imperative.

A recent study has reported that adding nanodiamond particles to graphene and sliding them

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against a diamond-like carbon (DLC) film-decorated counterpart ball enables macroscale superlubricity to be achieved in a dry nitrogen environment [21]. However, pure graphene does not exhibit a sufficiently superlow friction coefficient even though it does show excellent macroscale tribological performance [22–24]. It has been demonstrated that nanoparticles play a major role in achieving superlubricity [21]. Previous studies indicate that nanoparticles can effectively improve the tribological properties of various coatings [25–27]. In an intricate macroscale tribo-system with incorporated nanoparticles, the nanoparticles may have a roll effect and/or provide supporting action during the friction process [28]. Therefore, obtaining greater insight into the role of nanoparticles would help in better understanding the macroscale lubricating mechanism of novel tribo-materials such as superlubricating graphene/nanoparticle composites. In addition, the newly emerging graphene-nanoscrolls have attracted extensive attention due to their fascinating properties [29–31], particularly in superlubricating graphene/nanodiamond composites. Graphene nanoscrolls can be formed not only by graphene nanosheet-wrapped nanoparticles under shear force but also by graphene nanosheets themselves [21]. In this sense, revealing the relationship between graphene-nanoscroll formation and superlubricating performance is critical with respect to the effects of the structural parameters of nanoparticles and sliding parameters such as applied load. In addition, if a steel ball can be employed as a universal counterpart of superlubricating graphene/nanoparticle composites, the as simplified tribo-system would help in better understanding the lubrication mechanism and macroscale superlubricity performance of nanocomposites and also promote the application of the system. In this manner, the exploration of superlubricity in relation to the indispensable structural factors (e.g., nanoparticles and counterpart ball of the tribo-pair) of friction-reducing graphene/nanoparticle composites will shed light on reducing the friction and wear of mechanical moving parts [32, 33].

In this study, commonly used nano-SiO₂ particles are added to graphene nanosheets to obtain a graphene/nano-SiO₂ particle composite coating, and the tribological properties of the as obtained composite coating are compared with those of pure graphene coating.

First, different loads are applied to manipulate the formation of graphene nanoscrolls under the action of shear force in the presence of nano-SiO₂ particles as seeds, and graphene nanoscroll formation is investigated with respect to the superlubricating performance of the composite coating. Second, solid-structured spherical nano-SiO₂ particles and hollow-structured fullerenes with grapheme nanoscroll structures [34] are chosen to clarify the influences of the structural parameters of nanoparticles during the friction process. In addition, a widely used GCr15 counterpart steel ball is chosen to investigate whether the introduction of a DLC film-coated steel ball is essential for achieving superlubricity. Focusing on the roles and the structural parameters of the nanoparticles, this study reports the macroscale tribological properties of graphene/nano-SiO₂ particle composite coatings upon sliding against steel balls in terms of superlubricating material design and the practical use of superlubricity.

2 Experimental

2.1 Materials

Few-layer graphene (approximately 3–4 layers) powders were purchased from Jiangsu XFNANO Materials Technology Co., Ltd. (Nanjing, China); their original structure and morphology are shown in the Figs. S1 and S2 in the Electronic Supplementary Material (ESM). Fullerene nanoparticles (~20 nm) were purchased from Nanjing Jicang Nano Technology Co., Ltd. (Nanjing, China). Nano-SiO₂ particles (~20 nm) were purchased from Henan University (Kaifeng, China). Analytical grade ethanol (Tianjin, China) without any volatile agents was commercially obtained. All the as received materials were used directly without treatment.

2.2 Preparation of samples

Three portions of the same pure graphene powder were separately dispersed in ethanol to obtain dispersions. Then, graphene, nano-SiO₂ particles, and fullerene nanoparticle powder were added to the as obtained dispersions, and the mass ratio of the graphene and powder was 1:2. The resultant mixed

dispersions with a mass concentration of $100 \text{ mg}\cdot\text{L}^{-1}$ were sonicated for 1 h and then sprayed onto monocrystalline silicon (R_a : 2–3 nm) substrates. In the sample preparation process, the dispersions of graphene and/or nano-SiO₂ particles (or fullerene nanoparticles) were mixed without any binders or surfactants. In addition, high-purity nitrogen was used as the carrier gas and the pressure was maintained at 0.2 MPa during the entire spraying process. The as sprayed coatings without any contaminants were kept in a dry nitrogen atmosphere to prevent oxidation.

2.3 Tribological tests

Friction tests were conducted using a high vacuum ball-on-disk tribometer (CSM, Switzerland), where the atmospheric environment of the chamber was controllable. Prior to the sliding tests, the as prepared coatings were dried at 80 °C in vacuum ($\leq 0.1 \text{ Pa}$) for 1 h to evaporate residual agents, if any. The as dried coatings were paired with a counterpart steel ball (GCr15, diameter: 6 mm, R_a : 20 nm) and driven to rotate at room temperature ($23 \pm 2 \text{ }^\circ\text{C}$) in a dry nitrogen atmosphere (800 mbar) under applied loads from 0.5 to 3.0 N and a rotary speed of 60 r/min (radius of rotation: 4–6 mm). The friction coefficient curves were recorded for up to 1,800 cycles (30 min), during which the friction coefficients remained stable. The tribometer was calibrated twice automatically prior to each sliding test, and all friction tests were repeated under the same conditions at least three times.

2.4 Characterizations and measurements

The phase ingredients of the original graphene were identified by X-ray diffraction (XRD, Bruker, D8Discover25, Germany; copper target, $\lambda = 0.154 \text{ nm}$; tube current, 40 mA; tube voltage, 40 mV). The structure of the as received graphene powder was analyzed by a Raman spectroscope (Horiba Jobin Yvon S.A.S, LabRAM HR Evolution, France) at a laser wavelength of 532 nm. In addition, the as received powders (graphene, SiO₂, and fullerene nanoparticles) were dispersed separately in ethanol and ultrasonically treated for approximately 30 min. The as obtained dispersions were then transferred onto a Cu grid and dried in air to obtain samples for transmission electron microscopy (TEM) and high-resolution transmission

electron microscopy (HRTEM). The sliding interfaces after friction for approximately 30 min were also transferred to the Cu grids and used to analyze the microstructures and morphologies by the TEM and HRTEM (TF20, FEI, USA). The surface topographies of the as prepared coatings were characterized using the field emission electron microscope (JSM-6701F, Japan).

3 Results and discussion

3.1 Effect of nano-SiO₂ particles on the formation of graphene nanoscrolls

The tribological performances of graphene and graphene/nano-SiO₂ composite coatings under different applied loads were investigated to reveal the influence of nano-SiO₂ particles on the formation of graphene nanoscrolls and the relation between graphene-nanoscroll formation and superlubricating performance. Figure 1 schematically shows the procedures for preparing the graphene/nano-SiO₂ composite coating and the assembly of the frictional pair. The inset in Fig. 1(b) shows that nano-SiO₂ particles are well deposited on graphene nanosheets to enable a graphene/nano-SiO₂ composite coating, and the nano-SiO₂ particles are prone to agglomeration in the composite coating due to their high surface energy [35]. The other coating preparation and friction test processes were the same as the aforementioned.

The friction coefficient curves of the graphene/nano-SiO₂ particle composite coating and pure graphene coating under different applied loads are shown in Fig. 2. The graphene/nano-SiO₂ composite coating provided a friction coefficient of approximately 0.06 under an applied load of 0.5 N, and its friction coefficient declined to 0.03 at 1.0 N. As the applied load increased to 2.0 N, the graphene/nano-SiO₂ composite coating achieved superlubricity (the friction coefficient drastically declined to 0.006–0.008 (Fig. 2(a)). Macroscale superlubricity (friction coefficient of 0.010–0.011) of the graphene/nano-SiO₂ particle composite coating under a relatively high load of 3.0 N could also be obtained, but the lubricating coating will wear out quickly and the friction coefficient will increase steeply. This could be attributed to the deformation of the graphene nanosheets under high contact stress [36].

Fig. 1 Schematic of (a) graphene/nano-SiO₂ particle composite coating preparation and (b) friction pair assembly.

Fig. 2 Friction coefficient curves of (a) graphene/nano-SiO₂ composite coating and (b) pure graphene coating under different applied loads.

In addition, the graphene/nano-SiO₂ composite coating underwent a shortened running-in period as the applied load increased, which might be because normal contact stress benefits the formation of transferred film on the counterpart steel ball. In contrast to the graphene/nano-SiO₂ particle composite coating, the pure graphene coating could not achieve superlubricity under all applied loads, although it did show a friction-reducing ability (Fig. 2(b)). Therefore, it can be demonstrated that nano-SiO₂ particles play a critical role in achieving superlubricity of a graphene/nano-SiO₂ particle coating under a moderate applied load. In addition, the friction coefficients of pure graphene and graphene/nano-SiO₂ particles composite coatings under a low applied load of 0.5 N were shown to be 0.09–0.11 and 0.06–0.07, respectively, which indicates that the addition of nano-SiO₂ particles into graphene nanosheets is favorable to reducing friction even at a low contact pressure. This may be because nano-SiO₂ particles can be incorporated into the

composite coating and act as ball bearings under normal and shear forces to exert a rolling effect [37]. Nevertheless, as the present study showed, unless the applied load was sufficiently high, the graphene nanosheets could not encapsulate the rolling nano-SiO₂ particles to form graphene nanoscrolls and achieve superlubricity. Simultaneously, the pure graphene coating did not form graphene nanoscrolls upon sliding against the counterpart steel ball under either a high or low load, whereas an ordered lamellar slipping structure emerged on its sliding interface (Fig. S3 in the ESM). This indicates that pure graphene and graphene/nano-SiO₂ particle composite coatings operate under different lubricating mechanisms. Furthermore, even though the counterpart GCr15 steel ball is not coated by the DLC film, the graphene/nano-SiO₂ particle composite coating could still achieve superlubricity under a properly tuned applied load, which may be critical to the practical use of superlubricity in a simplified manner.

Figure 3 shows the TEM and HRTEM morphologies of the sliding interfaces of a graphene/nano-SiO₂ particle composite coating after 30-min friction under different applied loads. The graphene/nano-SiO₂ particle composite coating underwent insignificant change after the 30-min friction process under an applied load of 0.5 N, which corresponded to its lack of superlubricity in conjunction with the random distribution of nano-SiO₂ particles on the graphene nanosheets (Figs. 3(a) and 3(c)). It achieved superlubricity after sliding under an applied load of 2.0 N, which corresponded to the formation of graphene nanoscrolls with an interlayer spacing of approximately 0.38 nm (close to the *d* spacing of graphene; see Figs. 3(b) and 3(d)). The as formed graphene nanoscrolls had approximately 20 layers, which is more than 3–4 layers than in the original graphene (Fig. S1 in the ESM), suggesting that graphene nanoscrolls were formed in a layer-by-layer rolling of graphene nanosheets. In other words, the high applied load (shear force) promoted the interaction of graphene nanosheets and nano-SiO₂ particles in the graphene/nano-SiO₂ particle composite coating. Under the action of a normal load and shear force, the graphene nanosheets could encapsulate nano-SiO₂ particles layer by layer to form multilayer graphene nanoscrolls at the sliding contact interface, thus establishing a structural prerequisite

for achieving superlubricity during the friction process. Moreover, the *d* spacing of the formed graphene nanoscrolls (0.38 nm) was greater than the interlayer spacing of pure graphene (0.34 nm) [34], which may have been due to the tensile stress in the outer layers. As verified in previous studies, the increase in the interlayer spacing of graphene is favorable to reducing friction to a certain degree [38, 39], which could well account for the desired friction-reducing effect of the as formed graphene nanoscrolls. Because the formation of graphene nanoscrolls at an increased load (referring to a high contact stress) is essential for achieving macroscale superlubricity of the as prepared graphene/nano-SiO₂ particle composite coating, it can be inferred that the superlubricating graphene/nano-SiO₂ particle composite coating could be applicable to instrumental components working under harsh conditions (e.g., high contact pressure).

3.2 Load-carrying capacity of nano-SiO₂ particles

Because the graphene/nano-SiO₂ particle composite coating can achieve superlubricity upon sliding against the counterpart steel ball only when graphene nanoscrolls are formed, we wonder whether a composite coating directly made of graphene nanoscrolls could exhibit superlubricity under the same sliding conditions. To investigate this issue, hollow-structured fullerene

Fig. 3 TEM and HRTEM morphologies of the sliding interfaces of a graphene/nano-SiO₂ composite coating after 30-min friction under (a, c) a low applied load (0.5 N) without superlubricity and (b, d) under a high applied load (2.0 N) with superlubricity, respectively.

nanoparticles with a graphene-nanoscroll structure [34] were combined with graphene nanosheets to afford a graphene/fullerene nanoparticle composite. TEM images of the hollow fullerene nanoparticles and spherical nano-SiO₂ particles are shown in Fig. 4. Figure 4 shows that the sizes of the solid-structured SiO₂ nanoparticles (Fig. 4(a)) and hollow-structured fullerene nanoparticles (Fig. 4(b)) used were both approximately 20 nm.

The friction coefficient curves of the graphene/nano-SiO₂ particle composite coating, graphene/fullerene nanoparticle composite coating, and pure graphene coating under low (0.5 N) and high (2.0 N) loads are shown in Fig. 5. Under a low load, the friction coefficient of the graphene/nano-SiO₂ particle composite coating was 0.06–0.07, which was lower than that of the pure graphene coating (0.09–0.11), and the friction coefficient of the graphene/fullerene nanoparticle composite coating was 0.12–0.15. Under a high load, the pure graphene and graphene/fullerene nanoparticle composite coatings had high friction coefficients of 0.26 and 0.21–0.30, respectively, and the graphene/nano-SiO₂ particle composite coating had a low friction coefficient of 0.006–0.008, thus

achieving macroscale superlubricity. This indicates that the addition of hollow-structured fullerene nanoparticles to graphene did not contribute to a friction-reducing effect, whereas solid-structured nano-SiO₂ particles were indispensable in reducing the friction of the graphene/nano-SiO₂ particle composite coating under both low and high loads.

Figure 6 shows HRTEM morphologies of the sliding interfaces of the graphene/fullerene nanoparticle composite coating after 30-min friction under applied loads of 0.5 and 2.0 N. The hollow-structured fullerene nanoparticles became elliptical after sliding against the steel ball under 0.5 N (Fig. 6(a)), and the number of layers was close to that of the original fullerene nanoparticles. This means that the hollow-structured fullerene nanoparticles underwent a slight structural deformation after sliding under a low load (0.5 N). When the applied load increased to 2.0 N, the fullerene nanoparticles were nearly flattened into strips after sliding (Fig. 6(b)), and their number of layers was reduced to 2–4, which were considerably fewer than the number of 15–16 of the original fullerene nanoparticles (Fig. 4(b)). This was because a high contact force caused destructive structural deformation

Fig. 4 TEM morphologies of (a) nano-SiO₂ particles and (b) fullerene nanoparticles.

Fig. 5 Friction coefficient curves of the graphene/nano-SiO₂ composite, graphene/fullerene nanoparticle composite, and pure graphene coating under the applied loads of (a) 0.5 N and (b) 2.0 N.

Fig. 6 HRTEM morphologies of sliding interfaces of the graphene/fullerene nanoparticle composite coating after 30-min friction under applied loads of (a) 0.5 N and (b) 2.0 N.

and severe damage to the original hollow-structured fullerene nanoparticles. In other words, the hollow-structured fullerene nanoparticles could not support the macroscale applied load (even at a low applied force), and the original circular or spherical hollow nanoscrolls were prone to deformation under the action of normal and shear forces during the macroscale friction process. This may also explain why the friction coefficient of the graphene/fullerene nanoparticle composite coating was not lower than that of the pure graphene coating. By contrast, solid-structured nano-SiO₂ particles exhibited good load-carrying capacity and could support the macroscale applied force during the friction process. Thus, graphene/nano-SiO₂ particle composite coatings have a superlow friction coefficient and even exhibit superlubricity with the formation of graphene nanoscrolls under a proper applied load.

3.3 Lubricating mechanism of graphene/nano-SiO₂ particle composite coating

Figure 7(a) shows the assembly of the friction pair of the friction process of the graphene/nano-SiO₂ particle composite coating under a dry nitrogen atmosphere, where interferences of atmospheric oxygen and/or H₂O are eliminated. As Fig. 7(b) shows, the nano-SiO₂ particles are liable to deposition on graphene nanosheets and agglomeration because of their high surface energy in the as prepared composite coating. In addition, inevitable active chemical bonds exist on the surface of nano-SiO₂ particles, and these active chemical bonds may easily interact with the functional groups of the graphene nanosheets, thereby enhancing the interfacial interaction between the nano-SiO₂

particles and graphene nanosheets. During the friction process, the graphene nanosheets are deformed under the action of a high applied load (2.0 N), and the distance between graphene nanosheets and SiO₂ nanoparticles is reduced as their interaction is enhanced. At a load of 2.0 N, the normal force of the load provides sufficient pressure to the graphene/nano-SiO₂ particle composite coating, and the high applied load could afford sufficient shear force during the sliding friction process. Thus, a sufficiently high shear force enables the graphene nanosheets to wrap around the nano-SiO₂ particles. In other words, when the shear force is sufficiently high, it can promote scrolling (Fig. 7(c)) and then enable the solid-spherical nano-SiO₂ particles enwrapped by multilayer graphene nanoscrolls to reduce the friction through their function as ball bearings. The nano-SiO₂ particles are solid-structured spheres and can support the macroscale applied force in the graphene/nano-SiO₂ particle composite coating (Fig. 7(d)). During the friction process, the realization of superlubricity depends on the formation of graphene nanoscrolls at the sliding interface. In other words, the formation of graphene nanoscrolls is the structural prerequisite that enables the graphene/nano-SiO₂ particle composite coating to achieve superlubricity. Only when the formed graphene nanoscrolls are uniform and stable at the sliding contact interface can the friction coefficient be stable. Nevertheless, at the sliding interface, the existing graphene nanosheets and unchanged nano-SiO₂ particles can contribute to the fluctuation of the friction coefficient. As a result, the friction coefficient is unstable during the friction process (Fig. 2(a)). However, the hollow-structured fullerene nanoparticles with a graphene-nanoscroll structure cannot support the

Fig. 7 Schematics show superlubricating mechanism of graphene/nano-SiO₂ composite coating: (a) assembly of the friction pair of the friction process; (b) friction pair during the friction process and graphene/nano-SiO₂ particle composite coating; (c) role of nano-SiO₂ particles in promoting the formation of graphene nanoscrolls under the action of normal and shear forces; (d) solid-structured nano-SiO₂ particles play the load-bearing role to support the applied load.

macroscale force and are prone to deformation. Thus, the graphene/fullerene nanoparticle composite coating exhibits poorer friction-reducing ability than the graphene/nano-SiO₂ particle composite coating. In addition, along with the formation of graphene nanoscrolls, the contact area between neighboring graphene nanoscrolls or between graphene nanoscrolls and graphene nanosheets is reduced [28]. In other words, the contact area of the sliding interface is reduced in size when in the presence of solid-spherical nano-SiO₂ particles. Furthermore, the randomly distributed graphene nanosheets and graphene nanoscrolls may form incommensurate contact to lower the molecular potential energy [5, 6, 10, 11, 40, 41]. Moreover, the interactions of edges and dangling bonds of original graphene nanosheets are weakened along with the formation of closed-structure graphene nanoscrolls, which could lead to a decrease in atomic potential energy and friction force [42]. As a result, both the structural and sliding parameters function synergistically to reduce drastically the friction of the graphene/nano-SiO₂ particle composite coating and ultimately achieve macroscale superlubricity. With the progress of the friction test, the long-term friction

test process inevitably causes damage to all lubricating materials (graphene nanoscrolls, graphene nanosheets, and nano-SiO₂ particles), resulting in a collapse of materials (particularly the collapse of graphene nanoscrolls). Thus, with the destruction of the lubricating material of the sliding contact interface, the lubricating materials are gradually crushed and the nanocomposite coating is also gradually destroyed. Finally, the graphene/nano-SiO₂ particle composite coating wears out.

In summary, nano-SiO₂ particles can promote graphene nanosheets to form graphene nanoscrolls under a tuned applied load (2.0 N), which contributes to reducing friction. The applied load provides a normal force and promotes interaction between the graphene nanosheets and nano-SiO₂ particles in the composite coating. A shear force in the friction direction also exists at this time. Thus, the graphene nanosheets wrap the nanoparticles under the action of shear force and then form graphene nanoscrolls during the subsequent friction process. The added nano-SiO₂ particles can also help to support the macroscale force during the friction process, thereby helping to achieve macroscale superlubricity. In particular,

macroscale superlubricity can be exhibited by a simple system of a graphene/nano-SiO₂ particle composite coating sliding against a counterpart steel ball without DLC film decoration under proper conditions. This could be of significance for the application of macroscale superlubricity.

4 Conclusions

In this study, graphene/nanoparticle composite coatings were prepared on a monocrystalline silicon substrate. Macroscale friction tests showed that the graphene/nano-SiO₂ particle composite coating exhibited macroscale superlubricity (friction coefficient: 0.006–0.008) when sliding against a counterpart steel ball under an applied load of 2.0 N. This occurred because the incorporated nano-SiO₂ particles functioned as seeds to promote the formation of graphene nanoscrolls during sliding. The solid-structured nano-SiO₂ particles exhibited good load-carrying capacity and could support macroscale forces (normal load and shear force), both contributing to a drastic reduction in friction. This work clarified the two roles of nanoparticles in promoting the formation of graphene nanoscrolls and supporting macroscale forces for achieving macroscale superlubricity. In addition, the attainment of macroscale superlubricity was shown to be independent of the decorating counterpart steel ball with a DLC film, which could be meaningful for the design and application of graphene-based nanomaterials possessing ultralow friction.

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