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Laser irradiation-induced laminated graphene/MoS₂ composites with synergistically improved tribological properties

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Abstract

Engineering lubricant additives that have extraordinary friction reduction and anti-wear performance is critical to almost any modern mechanical machines. Here, we demonstrate the fabrication of laminated lubricant additives that can combine the advantages of zero-dimensional nanospheres and two-dimensional nanosheets. A simple *in situ* laser irradiation method is developed to prepare the laminated composite structure composed of ideally ultrasmooth MoS₂ sub-microspheres embedded within multiple layers of graphene. These ultrasmooth MoS₂ spheres within the laminated structure can change sliding friction into rolling friction under strong shear force created by moving contact surfaces to significantly reduce the friction. Meantime, the graphene layers can behave as 'protection pads' to efficiently avoid the formation of scars on the metal-to-metal contact surfaces. Overall, the laminated composites as lubricant additives synergistically improve the friction reduction and anti-wear properties. Additionally, due to the unique loosely packed laminated structure, the composites can stably disperse in the lubricant for more than 15 d and work under high temperatures without being oxidized. Such constructed laminated composites with outstanding tribological properties by an *in situ* laser irradiation method supply a new concept in designing lubricant additives that can combine the advantages of 0D and 2D structures.

Supplementary material for this article is available [online](#)

Keywords: 0D/2D laminated structure, laser irradiation, lubricant additives, tribological property

(Some figures may appear in colour only in the online journal)

1. Introduction

Friction is ubiquitous in nature, which wasted nearly 1/3 of human beings' consumed energy every year [1]. Therefore, reducing friction is critical to save energy and alleviate

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environmental pollution induced by the fossil energy consumption. Various approaches have been developed to minimize friction [2–4]. Among them, applying lubricants between contacting surfaces is probably one of the most successful strategies [5–7]. Recently, introducing micro/nanoscale additives into the lubricants has been proven to be able to further improve their friction reduction performance [8–12]. Excellent lubricant additives should have appropriate size, morphology, structure, and stiffness, as well as high stability and good dispersity in the lubricants. On one hand, zero-dimensional (0D) spherical particles can roll between contacting surfaces to reduce friction when used as lubricant additives [9–12]. On the other hand, two-dimensional (2D) layered materials as lubricant additives can behave as ‘protection pads’ between contacting surfaces to significantly enhance the anti-wear performance of the lubricant [13–15]. Therefore, 0D/2D composite structures may generate lubricant additives with outstanding tribological (i.e., friction reduction and anti-wear) performance. However, what is the favored organization way of the 0D and 2D structures to achieve the best tribological performance and how to experimentally realize it are still elusive [5].

On one hand, graphite and molybdenum disulfide (MoS_2), as two typical layered materials, have been widely studied as effective surface lubricants due to weak interatomic interactions and low strength shearing between interlayers [6, 16, 17]. The dispersion of pristine graphene and MoS_2 nanosheets in lubricating oil is usually unstable due to their high specific surface area. Serious aggregation and precipitation of graphene and MoS_2 nanosheets will result in unstable tribological performance [5, 7, 18–20]. On the other hand, MoS_2 spheres exhibit improved tribological behavior compared to the micro/nanoscale slices due to the better chemical stability and the spherical shape [8, 10, 11, 20]. However, fabrication of MoS_2 spheres requires high temperature (usually $> 900^\circ\text{C}$), high purity expensive and sometimes toxic gas precursors ($\text{H}_2\text{S}/\text{H}_2$), and harsh experimental conditions, which makes it unrealistic to employ MoS_2 spheres as lubricant additives [8, 21].

Here, we demonstrate a simple and one-step laser irradiation method to construct laminated composite structures formed by ultrasmooth and perfectly MoS_2 sub-microspheres wrapped within multiple layers of reduced graphene oxide (L-rGO/ MoS_2), which can effectively combine the advantages of 0D and 2D structures when used as lubricant additives (figure 1). These perfectly spherical MoS_2 particles can substantially transform sliding friction into rolling friction between two neighboring graphene layers acting as the ‘protection pads’ to simultaneously reduce the friction and enhance the anti-wear properties, effectively combining the advantages of 0D and 2D structures as lubricant additives. Importantly, the L-rGO/ MoS_2 structure can be well-dispersed in the lubricating oil without showing obvious precipitants for 15 d. Moreover, such composites can work under high temperatures without being oxidized. Therefore, the L-rGO/ MoS_2 structure can synergistically improve the tribological performance of the lubricating oil as lubricant additives. A simple one-step method is developed to prepare the L-rGO/ MoS_2

composite structures by laser irradiation of a dispersion composed of graphene oxide (GO) and MoS_2 nanoflakes. The MoS_2 nanoflakes are reshaped into ultrasmooth spheres and simultaneously are wrapped within the adjacent graphene nanosheets produced by laser-reduction of GO under the high temperature created by the laser irradiation. To the best of our knowledge, this is the first report of one-step fabrication of L-rGO/ MoS_2 composite structures that can integrate the advantages of 0D and 2D materials when used as lubricant additives, which opens up a new avenue towards engineering high-performance lubricant additives.

2. Experimental section

2.1. Preparation of the L-rGO/ MoS_2 composite structure

MoS_2 nanoflakes were synthesized by a hydrothermal method. Typically, 0.5 g sodium molybdate dehydrate (99.0% purity, Aladdin) and 0.8 g thioacetamide (99.0% purity, Aladdin) were added into 50 ml deionized water. After being stirred for 30 min, the solution was transferred into a 100 ml Teflon-lined stainless steel autoclave and heated at 200°C for 24 h. Then, the autoclave was cooled instantly to room temperature by a cold water bath. The MoS_2 flakes (figure S1(a) is available online at stacks.iop.org/NANO/29/265704/mmedia) were collected by centrifugation and washed several times with deionized water. GO nanosheets (figure S2(a)) were prepared by exfoliation of graphite (99.8% purity, 325 mesh, Alfa Aesar) according to a modified Hummers method [22].

The L-rGO/ MoS_2 composite structure was fabricated by pulse laser irradiation of a water dispersion of GO and MoS_2 at ambient conditions. 100 mg of the above-prepared MoS_2 flakes was added into 15 ml of a homogeneous GO dispersion (1.5 mg ml^{-1}) (referred as GO/ MoS_2), followed by stirring for 30 min to form a colloidal solution. A KrF excimer laser (10 Hz, 25 ns, Coherent, CompexPro 205) with a wavelength of 248 nm was used as the light source. The laser beam was focused into an area of around 0.87 cm^2 ($1.1\text{ cm} \times 0.79\text{ cm}$) at the solution through a convex lens with a focal length of 150 mm. The laser irradiation of the GO/ MoS_2 dispersion was lasted for different times at an energy fluence of $400\text{ mJ pulse}^{-1}\text{ cm}^{-1}$. The GO/ MoS_2 dispersion was continuously magnetically stirred during laser irradiation to make sure uniform irradiation and prevent sedimentation formation. After laser irradiation, the 0D/2D L-rGO/ MoS_2 particles were frozen dried to obtain powder products for further characterizations. Reduced graphene oxide (rGO) nanosheets and MoS_2 spherical sub-microspheres were obtained by laser irradiation of dispersions of pure GO and MoS_2 nanoflakes, respectively (figures S1(b) and S2(b)).

2.2. Material characterizations

The morphology of different samples was observed with a scanning electron microscope (SEM, FEI Quanta 250 FEG). The x-ray diffraction (XRD) pattern was obtained with an XRD apparatus (D8-Advance, Bruker) operated at 40 kV and 40 mA using the Cu- $\text{K}\alpha$ line ($\lambda = 0.154\text{ 184 nm}$) as the

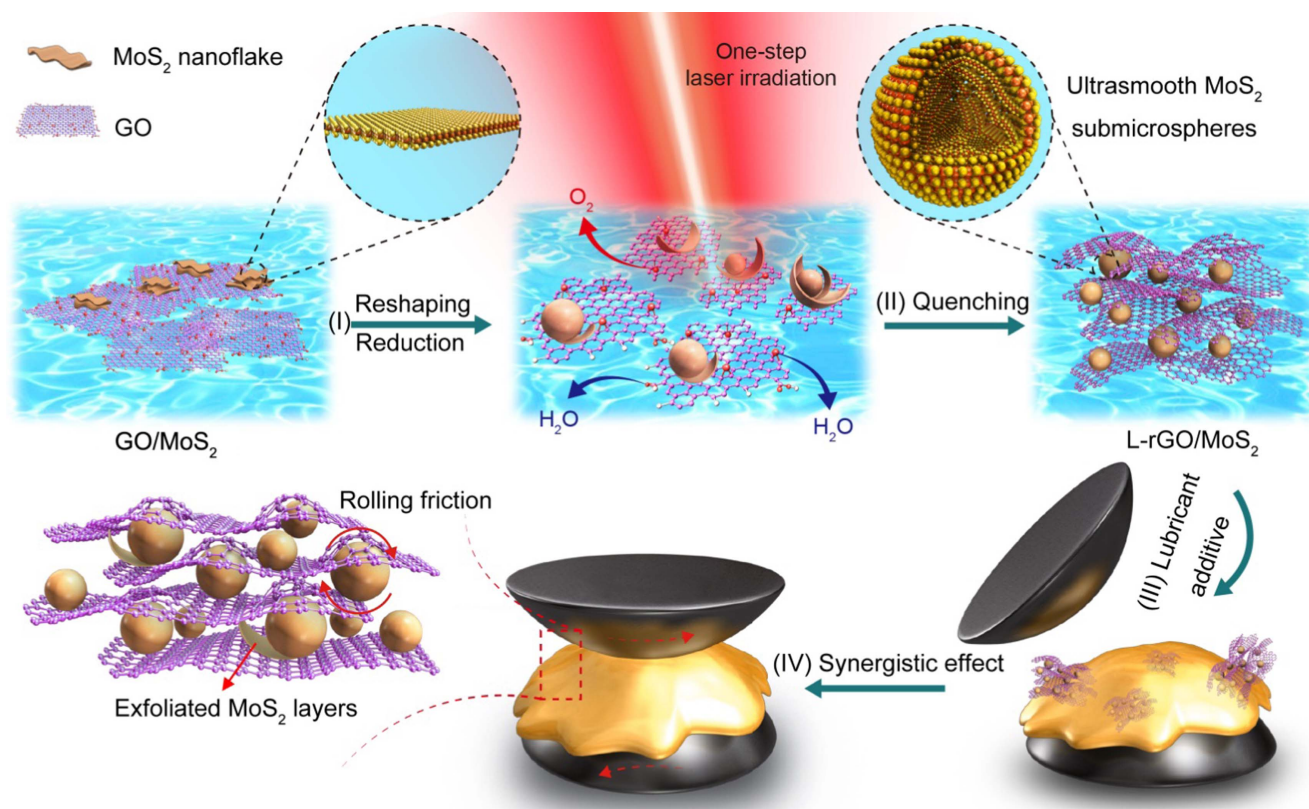


Figure 1. Schematic of the formation process of the L-rGO/MoS₂ composite structures and the working mechanism of friction reduction and anti-wear when they are used as lubricant additives. Process I. Pulsed laser irradiation of GO/MoS₂ creates localized high temperature and pressure, which induce reshaping of MoS₂ and reduction of GO to rGO. Process II. The quenching process of the surrounding liquid medium ‘freezes’ and solidifies the liquid MoS₂ droplet into ideally spherical particles. Several graphene layers wrap a tremendous amount of MoS₂ spheres, giving rise to the formation of L-rGO/MoS₂ composite structure. Process III. The L-rGO/MoS₂ composite structures are introduced into engine oil as lubricant additives. Process IV. The unique L-rGO/MoS₂ composite structure can combine the advantages of quasi-fullerene-like MoS₂ spheres and graphene nanosheets regarding friction reduction and anti-wear.

excitation source. Microstructural examination was characterized with a transmission electron microscope (TEM, JEM-2100F) under a 200 kV acceleration voltage. Raman spectrometer equipped with a 532 nm laser (LabRAM HR Evolution, HORIBA) was used for recording the Raman scattering spectra of different samples. The UV–visible absorption spectra were measured with a Shimadzu UV-3600 spectrophotometer (Shimadzu). The morphology of wear surface after four-ball test was observed with a laser scanning confocal microscope (LSCM, KEYENCE Corporation, VK-X250K). The binding energies of Mo in different samples were detected by x-ray photoelectron spectroscopy (XPS, Thermo Fisher, with an Al K α x-ray source).

2.3. Evaluation of the tribological properties

A four-ball tribology tester (MM-W1B, Shijin-Jinan, China) was used to study the tribological properties. The tester was operated with one steel ball under load f rotating at a speed of ω against three steel balls held stationary in the form of a cradle which is controlled by a DC servo motor. The steel balls with a hardness of 64–66 HRC were selected according to the National Standard of China (G20, GB/T308-2002). The 10w-40 engine oil was used as a reference lubricant in the

tribology test. The kinematic viscosity of the reference oil is $122 \text{ mm}^2 \text{ s}^{-1}$ at 40°C . Lubricant additives were added into the base oil at a concentration of 0.2 wt%, which were generally ultrasonically agitated for 30 min. Due to the poor dispersity, the GO/MoS₂ hybrids were sonicated for 2 h. The rotation speed of the rolling ball is 1200 rpm under a load of 392 N. The duration time is 30 min. The tests were performed at room temperature unless otherwise specified. Friction reduction was recorded automatically with a computer controlled data acquisition card. Wear scar diameter (WSD) was measured with a metallographic microscope. After washing with petroleum ether and acetone, the size and morphology of the wear scar area were evaluated by SEM and digital stereomicroscope (VHX-5000, KEYENCE).

3. Results and discussion

3.1. Fabrication and characterization of the L-rGO/MoS₂ composites

A simple one-step laser irradiation of a water suspension composed of GO/MoS₂ is used to fabricate the L-rGO/MoS₂ composite structure (figure 1). Due to the ultrahigh pressure

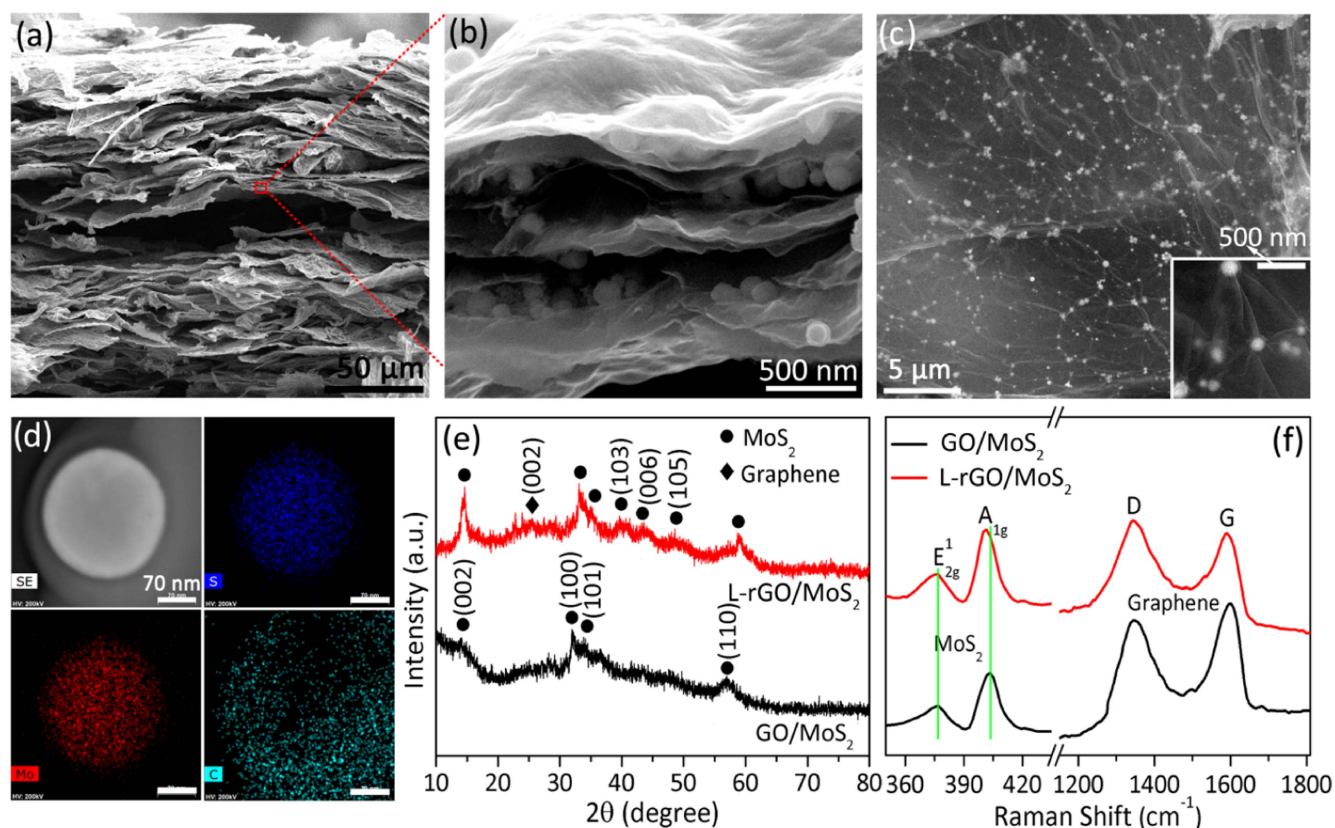


Figure 2. Scanning electron microscopy micrographs of the L-rGO/MoS₂ composite structure from (a), (b) the cross section and (c) the top view. (d) Element mapping of a MoS₂ sphere within the composite structure. (e) XRD results and (f) Raman spectra of raw GO/MoS₂ and L-rGO/MoS₂ composite structure.

(1 GPa) and ultrahigh temperature (10^4 K) around the laser spot-material interface induced by laser irradiation [23–29], MoS₂ nanoflakes are transformed into ideally spheres (figures 1 and S1) and meanwhile GO nanosheets are reduced to rGO with prominently reduced size (figures 1, S2, and S3). Interestingly, MoS₂ spheres are wrapped within graphene layers, forming the L-rGO/MoS₂ composite structure (figures 2(a)–(c)). The number density of MoS₂ spheres on rGO nanosheets is about $7/\mu\text{m}^2$ obtained by directly counting from figure 2(c). The energy-dispersive x-ray spectroscopy elemental mapping results indicate that the ultrasmooth MoS₂ sub-microspheres are perfectly wrapped by graphene nanosheets (figure 2(d)), which can effectively prevent the aggregation of rGO nanosheets by forming graphene wrinkles and the pulverization of MoS₂ spheres during the high-load friction process as proved later (figure 1).

Representative XRD patterns of raw GO/MoS₂ hybrids and L-rGO/MoS₂ composites prove the formation of MoS₂ spheres with a hexagonal structure. The broad diffraction peak appearing at $\sim 26.5^\circ$ in L-rGO/MoS₂ composites can be indexed to the (002) disorderly stacked layer of rGO nanosheets, proving the laser-induced reduction of GO to rGO (figure 2(e)). The Raman spectrum shows that the in-plane E_{2g}^1 phonon mode of MoS₂ sphere slightly redshifts compared to that of the nanoflakes, induced by the morphological transition from nanoflakes to quasi-fullerene-like MoS₂ spheres as discussed later (figure 2(f)). The relative

peak intensity ratio of I_G/I_D of graphene decreases when compared with that of GO, which is caused by laser-induced distortion and fragmentation of GO (figures S2 and S3).

High-resolution transmission electron microscope characterization indicates that MoS₂ nanoflakes have a poor crystallinity (inset in figure 3(a)). After laser irradiation, MoS₂ nanoflakes are transformed into MoS₂ spheres (figure 3(b)). MoS₂ spheres possess a nearly closed-cage structure due to the partial bonds closing at the edge of spheres (figures 3(c), (d)). Compared with raw MoS₂ flakes, ultrasmooth MoS₂ sub-microspheres show better-resolved lattice fringes with an interplanar distance of 0.62–0.64 nm from the (002) planes. The external layers of the MoS₂ sub-microspheres show a 1.5%–3% dilation of interlayers spacing along the *c* axis. The expansion is generally due to the presence of residual stresses in the curved layers. Some edge dislocations can also be observed within the quasi-fullerene-like structure (see red circle-marked area in figures 3(c), (d)). The subtle redshift of A_{1g} phonon mode in the L-rGO/MoS₂ composites (figure 2(e)) is an indicator of these changes in the microstructure and the morphology. More importantly, MoS₂ spheres are wrapped by the graphene layers, instead of simply attaching on the graphene layers, which can be proved by the bond layer between MoS₂ and graphene (figures 3(d) and S4). Such interfacial bonds can stabilize the sulfide particles on the graphene support, which helps to prevent graphene and MoS₂

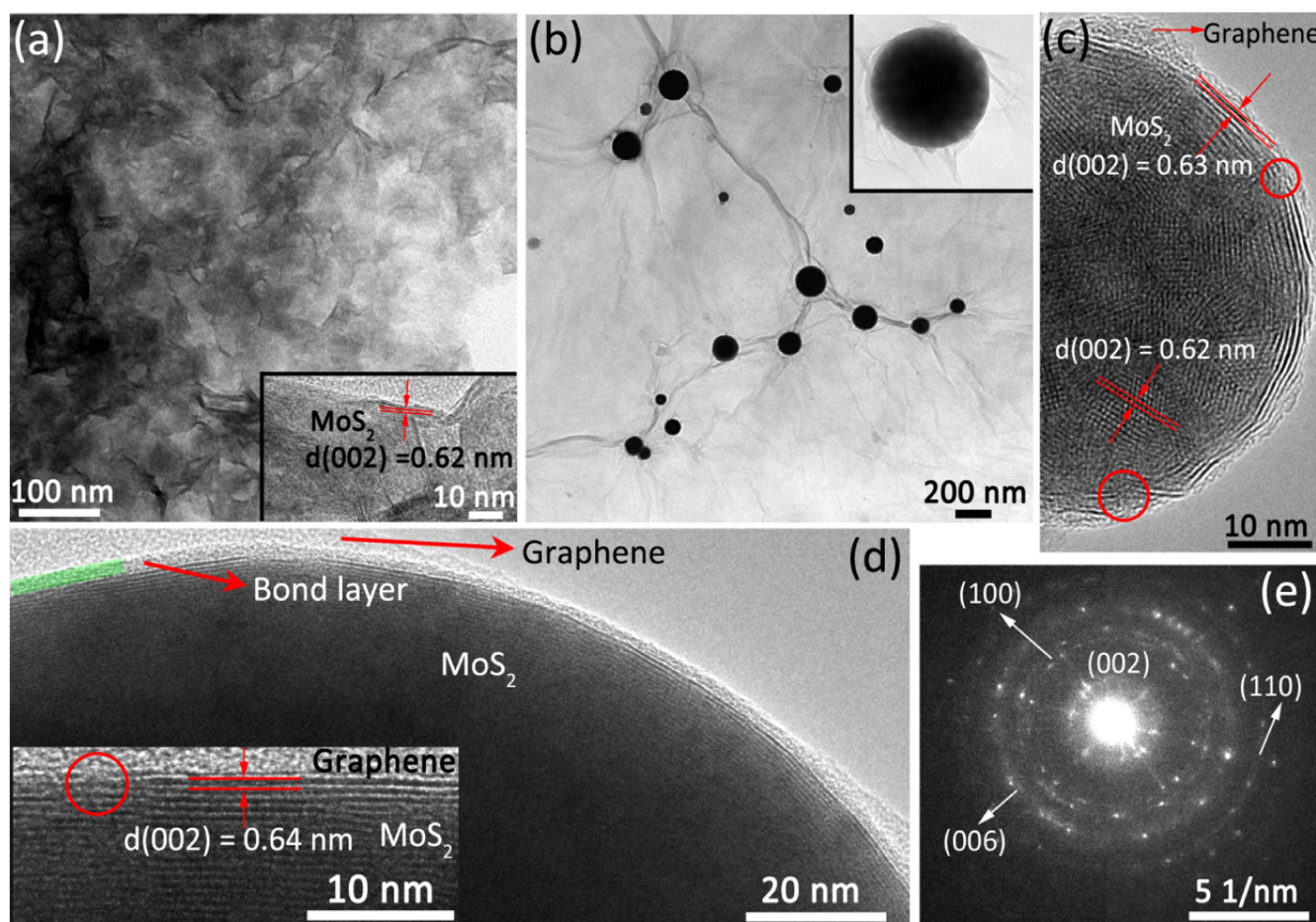


Figure 3. Microstructure analysis of the L-rGO/MoS₂ composite structure. TEM image of (a) raw GO/MoS₂ and (b) L-rGO/MoS₂ composite structure. (c)–(d) High-resolution TEM image of single MoS₂ spheres. (e) SAED pattern of the corresponding MoS₂ sphere.

from agglomeration and enables their good dispersion in the engine oil. The selected-area electron diffraction result reveals that the MoS₂ sub-microspheres are polycrystalline (figure 3(e)).

3.2. Formation mechanism of the L-rGO/MoS₂ composites

To systematically study the formation mechanism of the L-rGO/MoS₂ composite structures, we observed the shape evolution of the MoS₂ nanoflakes under laser irradiation for different times (figure S5). Laser irradiation of MoS₂ nanoflakes can create ultrahigh temperature and pressure within nanoseconds at a localized area [26, 27], which leads to bending and melting of the nanoflakes and subsequent evolution into spherical particles under successive laser pulse heating and liquid quenching processes (figure 1). Due to the large specific surface area of GO nanosheets, significant optical absorption below 350 nm could result in a rapid rise in temperature and pressure, which induced the transformation of GO to graphene due to the photothermal reduction effect [28, 29], as shown in figure S3. This transformation process is reflected by the color evolution of GO solution from the original yellow golden to bronzing and eventually to black color. Moreover, the C=O absorption peak of GO at 304 nm disappeared after laser irradiation. The absorption of GO at

230 nm redshifts to 256 nm, induced by the $\pi \rightarrow \pi^*$ transitions of extended aromatic C–C bonds after the KrF excimer laser irradiation [23, 29]. Overall, the deoxygenation of the GO nanosheets and the restoration of the sp^2 carbon sites in the graphene are confirmed.

The GO/MoS₂ hybrids possess layered structure with GO and MoS₂ nanoflakes stacking together due to their high specific surface energy [22]. Upon laser irradiation, the MoS₂ nanoflakes begin to bend and reshape into ultraspherical spheres within the stacked graphene layers (figure 4). Meantime, GO is reduced to rGO. Eventually, L-rGO/MoS₂ composite structures with a large interlamellar spacing are obtained (figure 4(d)). More interestingly, owing to the really high temperature around the MoS₂ spheres, the graphene layer is formed around the MoS₂ spheres (figures 3 and S4), which can maintain MoS₂ spheres inside the graphene layers even under strong shear force when used as lubricant additives as discussed later.

3.3. Tribological properties of L-rGO/MoS₂ composites as oil additives

The dispersity of the L-rGO/MoS₂ composite structures in the bare engine oil is first investigated (figure 5(a)). No observable precipitations appeared even after 15 d in the engine oil,

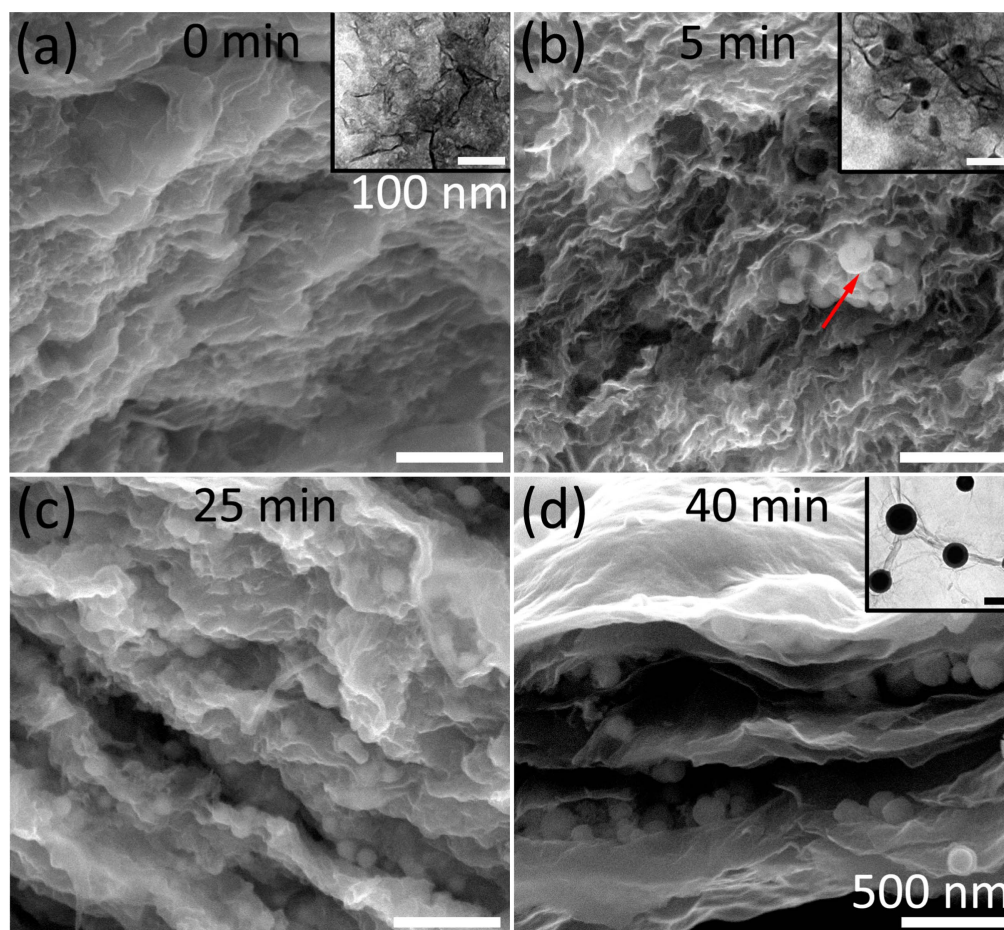


Figure 4. Formation process of the L-rGO/MoS₂ composite structure. Morphology evolution of the mixture of GO nanosheets and MoS₂ nanoflakes after laser irradiation for different times: (a) 0, (b) 5, (c) 25 and (d) 40 min. The laser fluency is 400 mJ pulse⁻¹ cm⁻¹. Inset: corresponding TEM images. The red arrows show the early stage formation of spherical particles.

demonstrating that the laminated composite structures could stably disperse in the engine oil. In contrast, obvious precipitations emerged in the solution composed of GO/MoS₂ after only 1 d. These results are consistent with the Zeta potential and the morphological measurements (figures S6 and S7). The absolute value of the Zeta potential of the solution composed of 0D/2D L-rGO/MoS₂ composite structures is much higher than that of the solution composed of GO/MoS₂. Due to the flat, disk-like shape and high specific surface energy of MoS₂ flakes and GO nanosheets, they are prone to aggregating together. These aggregates tend to precipitate in the engine oil. Oppositely, the crumpled surface of the L-rGO/MoS₂ composite structures prevents them from attacking each other and makes them stably disperse in engine oil [6, 30].

The tribological properties of the L-rGO/MoS₂ composites as engine oil additives at a concentration of 0.2 wt% (figure S8) were evaluated by a four-ball tribology tester (figure 5(b)). To compare, the same tribology tests were performed on the pure engine oil and the engine oil containing rGO nanosheets, MoS₂ nanoflakes, laser irradiation-created perfectly spherical MoS₂ particles, and GO/MoS₂ hybrids, respectively. During the first 5 min, the coefficients of friction (referred as COF) increase for all of the lubricants, which is because the contact between the balls transformed

from point contact to surface contact (figure 5(c)). All of the additives can reduce the COF after the friction coefficient is stabilized. Graphene additives can reduce the COF by about 6% when compared to that of the pure engine oil. The COF of MoS₂ nanoflake additives is on average comparable to that of the graphene additives, but increases gradually. By introducing GO/MoS₂ into the engine oil, the COF is reduced to a larger extent than only introducing one of the two. The laser irradiation produced ideally MoS₂ spheres as lubricant additives can effectively reduce the COF by ~17% when compared with that of the pure engine oil. This is because the ultraspherical particles can effectively turn sliding friction into the rolling friction [31, 32]. Strikingly, the L-rGO/MoS₂ composite structures can reduce the COF by ~25% when compared with that of the pure engine oil (figures 5(c) and (e)). Notably, the friction coefficient of the engine oil composed of 0D/2D L-rGO/MoS₂ composite structures fluctuates very slightly (<~2%) and does not show obvious increase during the measurement. In contrast, the friction coefficient of pristine engine oil and engine oil with other four additives all shows obvious fluctuations (>10%) (figure 5(b)). Additionally, the friction coefficient of the 0D/2D L-rGO/MoS₂ composites does not change too much even working under 75 °C (figure 5(e)). In contrast, the pure engine

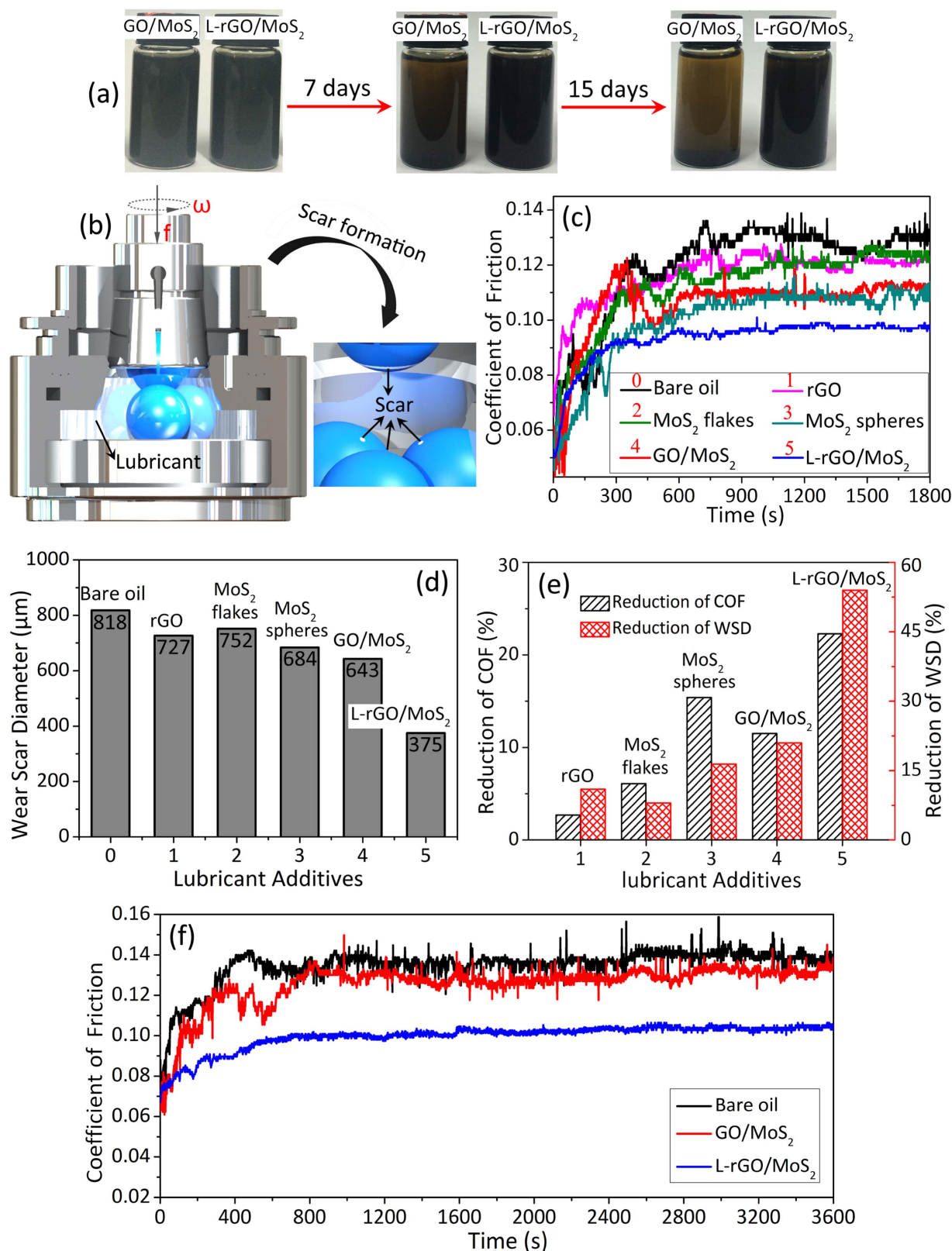


Figure 5. Friction reduction and anti-wear properties. (a) Dispersion properties of raw GO/MoS₂ hybrids and L-rGO/MoS₂ composites. (b) Schematic of the four-ball tribology tester for friction coefficient and wear scar measurements. (c) COF and (d) WSD of the engine oil with different additives (0.2 wt%) after running for 30 min at room temperature. (e) The corresponding reduction percentage of average COF and WSD compared with those of the bare engine oil. (f) COF of the bare oil and the oil contained GO/MoS₂ and L-rGO/MoS₂ composite structure for 60 min at 75 °C.

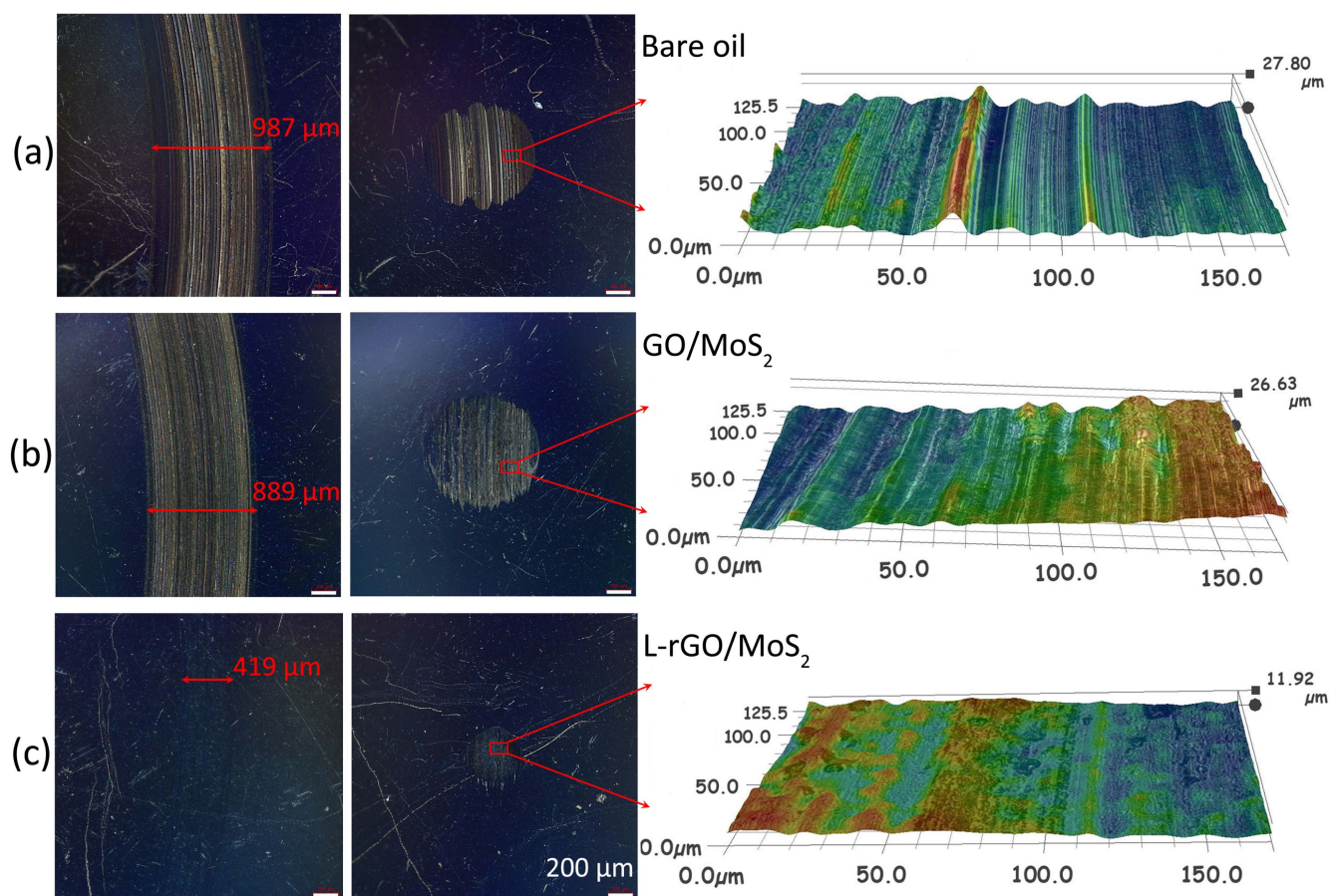


Figure 6. Optical images (left column) and the corresponding 3D profile images (right column) of the wear surfaces lubricated sliding with (a) bare oil, (b) raw GO/MoS₂ hybrids, and (c) L-rGO/MoS₂ composites for 60 min at 75 °C.

oil and the oil contained GO/MoS₂ increases prominently at high temperatures, owing to the rupture of boundary film at adhesion point.

Desirable lubricant additives should not only be able to reduce friction, but also have good anti-wear properties since serious wear will cause mechanical failure of the engine [33–36]. The anti-wear property is usually evaluated by the WSD on moving balls. All of the lubricant additives can reduce the WSD (figures 5(d), (e)). In order to clearly observe the wear scar, the top rotating ball and one stationary ball lubricated sliding with bare oil, raw GO/MoS₂ hybrids, and L-rGO/MoS₂ composites for 60 min at 75 °C, were characterized by a LSCM, as shown in figure 6. Raw GO/MoS₂ additives reduce the WSD on stationary ball by only ~10% when compared to that of the pure engine oil. In particular, introducing the L-rGO/MoS₂ composite structures into the engine oil, the WSD on stationary ball decreases from 987 to 419 μm (~58% reduction). The 3D maps of the wear area on stationary balls indicate the wear tracks are very shallow and small. The absence of deep and wide wear tracks can prevent the formation of large debris that can inflict severe abrasive wear [37, 38]. Therefore, no obvious wear tracks on the rotating ball are produced and the WSD decreases by about 58%, which outperforms most of the previously reported lubricant oil additives under such a high-load and rotation speed (table S1). Overall, the 0D/2D L-rGO/MoS₂

composite structures are excellent lubricant additives with outstanding dispersion stability in lubricating oil and friction reduction and excellent anti-wear tribological properties.

3.4. Working mechanism in friction reduction and anti-wear processes

To clarify the lubricating mechanism of the L-rGO/MoS₂ composite structure as lubricant additives, we analyzed the wear area composition after the tribology test with the Raman spectrometer and energy-dispersive x-ray spectrometer (EDS). In light of the discontinuity of tribofilm after repeated cleaning by petroleum ether, acetone and alcohol, we performed Raman intensity mapping over a larger area (100 × 75 μm) in the wear scar (the red box), as shown in figure 7(a). Raman intensity mapping images of the graphene G mode and the MoS₂ A_{1g} mode indicate the transfer of graphene and MoS₂ to metal friction surface. The existence of Mo and S elements (figure 7(b)) on the wear area also indicates the formation of tribofilm on wear surfaces by the 0D/2D L-rGO/MoS₂ composite structures. Due to the large specific area of graphene, the laminated composite structure has a great chance to be transferred into the metal-to-metal contact surface area during the ball movement to prevent metal-to-metal direct contact, acting as a spacer to prevent the scar formation [22, 39]. More importantly, the quasi-fullerene-like MoS₂ spheres between the graphene layers can change

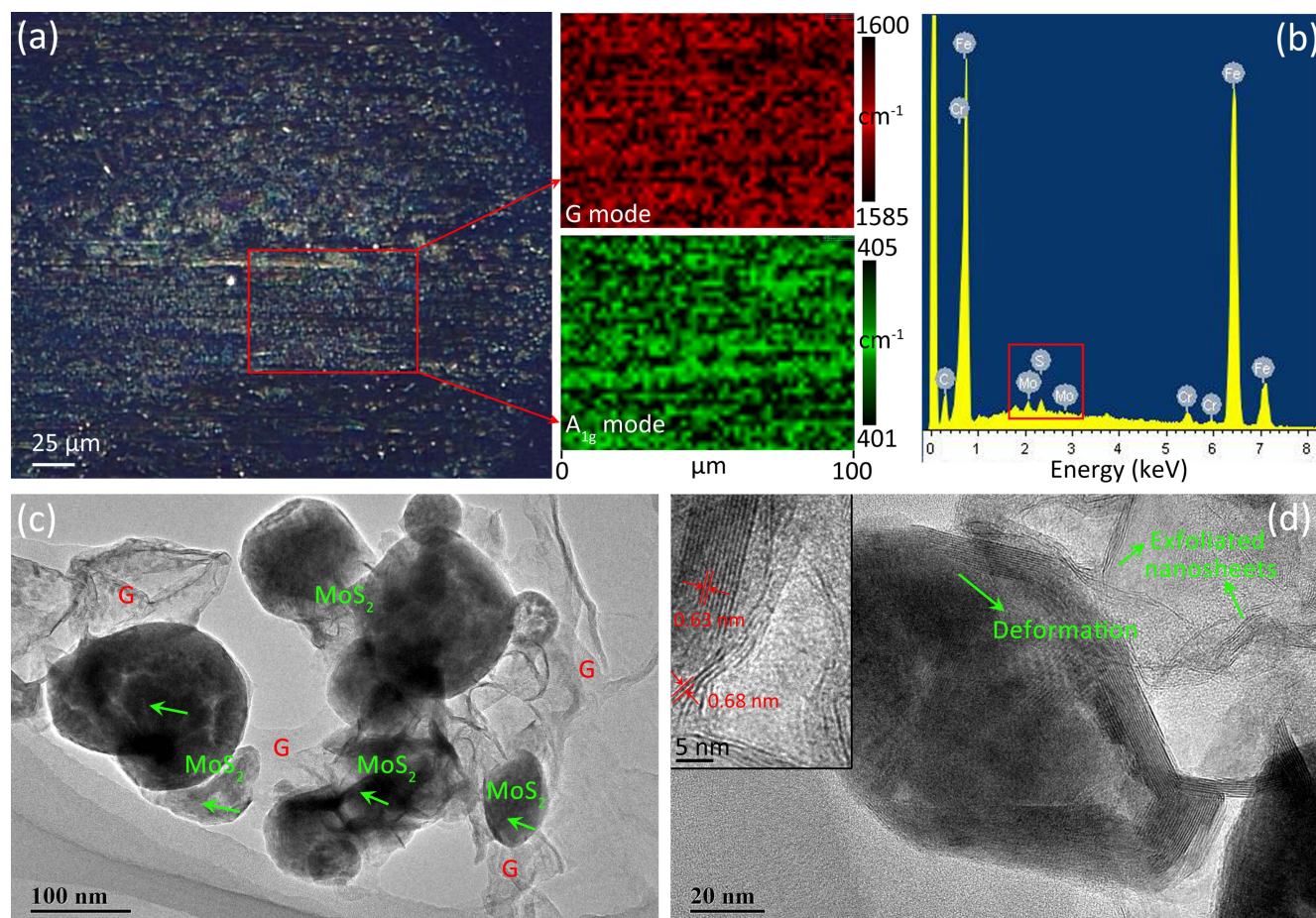


Figure 7. Working mechanism of the L-rGO/MoS₂ composites regarding friction reduction and anti-wear. (a) Raman intensity mapping images of graphene with the G mode (red) and MoS₂ with the A_{1g} mode (green). (b) EDS of the wear area lubricated by engine oil composed of L-rGO/MoS₂ composites for 60 min under 75 °C. (c) and (d) TEM images show the morphology and microstructure of L-rGO/MoS₂ composite structure after the tribology test. The deformation of MoS₂ nanospheres and exfoliation of nanosheets are observed.

sliding friction into rolling friction under the strong shear force caused by the moving balls, making the 0D/2D L-rGO/MoS₂ composite structures analogous to micro-bearings to greatly reduce the friction [31]. So, the micro-bearing spheres can be deformed or even exfoliated under high-load shear force, as shown in figures 7(c), (d). Then these exfoliated flake-like MoS₂ nanosheets as additives still protect the steel surfaces.

In addition, the raw MoS₂ nanoflakes are of highly chemical activity as there are many chemical dangling bonds at the edge sites. So, MoS₂ nanoflakes are easily oxidized to MoO_x, which is generally considered as a main reason for the invalidation of MoS₂ lubricants because of the damage of layered structure [31, 33]. The quasi-fullerene-like MoS₂ spheres are rare with dangling bonds, as well as they are protected by the circumjacent graphene layers, allowing L-rGO/MoS₂ composites to be used under high temperatures without being oxidized (figures 8 and S9).

4. Conclusions

Comprehensively considering the working mechanism of inorganic particles as lubricant additives, we construct a novel

laminated lubricant additives that can combine the advantages of 0D and 2D structured lubricant additives. A simple one-step method is developed to prepare the L-rGO/MoS₂ composite structure by laser irradiation of a dispersion composed of MoS₂ nanoflakes and GO nanosheets in ambient conditions. The high temperature and pressure created by the laser irradiation can reshape the MoS₂ nanoflakes into perfectly spherical particles with a quasi-fullerene-like structure and meantime reduce the GO into rGO. The laminated composite structures are composed of ultraspherical MoS₂ sub-microspheres embedded within multiple layers of graphene. The spherical MoS₂ particles can change sliding friction into rolling friction under strong shear force to effectively reduce the friction. At the same time, the 2D graphene layers as lubricant additives can protect the metal contacting surfaces from being scratched, significantly enhancing the anti-wear performance of the lubricant. Overall, the 0D/2D L-rGO/MoS₂ composites have outstanding tribological properties even under high working temperatures when used as lubricant additives, better than conventional nanoparticles and 2D materials. These characteristics endow them with great application potentials as high-performance practical lubricant additives.

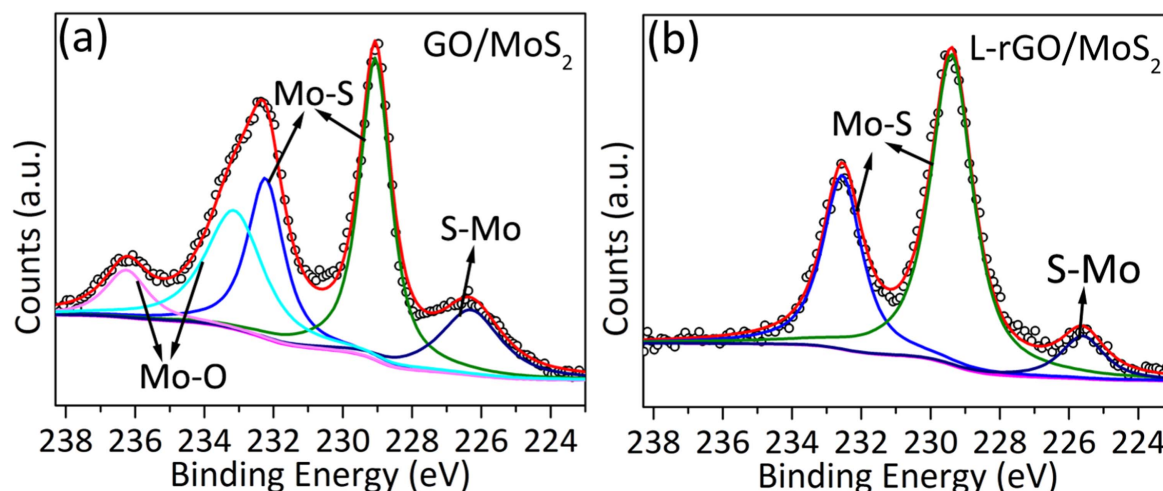


Figure 8. XPS spectra of (a) raw GO/MoS₂ hybrids and (b) L-rGO/MoS₂ composites used as lubricant additives after tribology test for 60 min under 75 °C.

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