

Multi-asperity micro-spherical interfaces enabling macroscale superlubricity under various atmospheric conditions

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Abstract: Superlubricity ($\mu < 0.01$) could revolutionize energy-efficient technologies but remains confined to nanoscale or controlled environments. Here, we propose a universal strategy that overcomes these barriers by incorporating numerous microspheres to form a self-assembled friction interface, transforming macroscopic surface contact into microscopic point contacts and inducing structural superlubricity at each contact, thereby achieving macroscale superlubricity. This is realized through the *in-situ* formation of hydrogenated fullerene-like carbon-coated nanospheres (HFLC)/MoS₂ tribofilm at frictional interface. Friction-induced embedding of HFLC nanospheres into MoS₂ films creates multi-asperity interfaces that convert macroscopic contact into numerous micro point contacts. This structure reduces the contact area, suppresses chemical adhesion, and endows the shear interface with homogeneous and heterogeneous incommensurate shear potential, thereby triggering macroscale superlubricity. The nanospheres retain their spherical geometry during operation, sustaining ultralow friction ($\mu = 0.009$) on steel substrates under alternating vacuum-air environments. Our strategy provides a scalable pathway to achieve near-zero friction in industrial systems.

Keywords: Macroscale superlubricity, heterojunction structure, carbon nanospheres, tribofilm

1 Introduction

Energy consumption due to friction accounts for more than a third of the world's primary energy, resulting in the failure of about 80% of mechanical components [1, 2]. Hence, the advancement and design of lubrication strategies with superior friction-reducing capabilities have become a key research focus [3-7]. The solid superlubricity significantly contributes to the reduction of friction and wear, it can be classified into microscale structural superlubricity and macroscale superlubricity, each with distinct advantages and limitations. Structural superlubricity represents a state of near-zero friction loss [2]. It has been found that this phenomenon is particularly significant when the two crystal surfaces are not commensurate [2].

Since the concept was first introduced by Shinjo and Hirano in the 1990s, significant progress has been made in the development of superlubricity technology. It appears to be the optimal lubrication strategy to address the energy waste caused by friction and wear. Research on two-dimensional (2D) materials, including graphene and transition metal dichalcogenides (TMDCs), has extensively confirmed superlubricity at the nanoscale and microscale [8]. Despite advances at smaller scales, achieving macroscale superlubricity remains challenging due to a variety of factors. The difficulty in producing large-sized 2D single-crystal materials and transferring them to engineering substrates nondestructively is a significant hurdle [9]. Additionally, the formation of ordered junctions in van der Waals (vdW) heterostructure nanopowders before friction and the management of complex contact reconstruction among numerous finite-sized connect post-formation of ordered frictional junctions pose further challenges. Technological constraints also make it difficult to produce continuous, defect-free macroscopic contact surfaces at the molecular level [10]. Hence, directly achieving near-zero friction and wear through structural superlubricity at the macroscale is challenging. However, if the microscale structural superlubricity can be extended to the macroscale, stable macroscale superlubricity can be realized. This superlubricity technology can significantly reduce energy loss and material wear during friction processes, enhancing the reliability and service life of moving

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components [11].

In recent years, researchers have discovered that by optimizing the surface structure and chemical composition of the contact surface, the friction coefficient can be significantly reduced, achieving a state of superlubricity [12-16]. For instance, the use of graphene and other 2D materials has demonstrated considerable potential for reducing friction at the nanoscale. These materials possess unique properties that enable the formation of nearly frictionless interfaces when correctly aligned [17]. Furthermore, the advancement of van der Waals (vdW) heterostructures has significantly enhanced the understanding and application of solid superlubricity. These heterostructures, consisting of stacked layers of various 2D materials, can foster favorable conditions for ultra-low friction by minimizing interlayer interactions [18]. The core of the aforementioned research is to develop a macroscopically continuous, defect-free (or uniformly oriented) tribofilm under friction. This novel macroscopic tribofilm achieves large-scale structural superlubricity at the macroscale. Beyond material selection, there are studies that bridge the gap between the macro and micro scales. For example, T. Ma et al. demonstrated that graphene-coated microspheres can provide an uneven multi-asperity contact, and their superlubricity is accompanied by high load-bearing capacity [19]. In addition, in the study by J. Li et al., such a contact mode with multiple microscopic contact points can provide conditions for homogeneous and heterogeneous shear, thereby achieving macroscopic superlubricity, especially under incommensurate contact conditions [20, 21]. By introducing or generating nano-coil structures within the tribofilm, these nano-coils, with their reduced contact area, can significantly lower the coefficient of friction [22]. In summary, by elaborately designing the microscopic morphology of the contact region, the macroscopic contact surface can be divided into microscale contact points. This reduces the contact area while minimizing the influence of macroscopic defects, thereby achieving an ideal lubrication effect [23]. Based on recent advances, we propose that the realization of macroscale superlubricity is essentially an extension of structural superlubricity at the microscale.

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Based on a summary of various macroscopic superlubricity strategies, this study proposes a novel method. Briefly, numerous hydrogenated fullerene-like carbon (HFLC) nanospheres are employed to transform the macroscopic single-asperity contact into a microscopic multi-asperity contact mode. In this contact configuration, the combination with MoS₂ films on steel substrates achieves environmental-insensitive macroscale superlubricity ($\mu = 0.009$).

2 Experimental

2.1 Materials preparation

Preparation of hydrogenated fullerene-like carbon coated nanospheres (HFLC nanospheres): SiO₂ nanospheres, averaging about 500 nm in diameter, were used as the initial substrate. HFLC nanospheres were prepared using PECVD technology. The carbon film was deposited using a gas mixture of argon/methane with a flow rate of 300/18 sccm, an applied bias voltage of -950 V at a pulsed frequency of 70 kHz with a duty cycle of 0.7, and a constant working pressure of 13 Pa.

Preparation of molybdenum disulfide (MoS₂) film: Select a 40×40×1.5 mm 304 stainless steel plate as the substrate. Use magnetron sputtering technology to deposit MoS₂ film on the stainless-steel plate. Before coating, the substrate must be ultrasonically cleaned in ethanol solution for 30 minutes. After cleaning, the cleaned steel plate is mounted in the sputtering chamber so that the polished surface (i.e., mirror surface) is exposed. The MoS₂ film is deposited on the 304 stainless steel substrates (with a polished surface) by using the equipment name. First, close the chamber and use a mechanical pump to evacuate the rough vacuum to 1.8 Pa. Then, use a molecular pump to evacuate the high vacuum to 1.8×10^{-4} Pa, and introduce Ar gas to adjust the internal pressure to 0.8 Pa. At the same time, adjust the RF power supply to 200 W, and keep the substrate temperature at 250 °C.

Combination of HFLC nanospheres and MoS₂ films: First, we mixed HFLC nanospheres with ethanol to form a mixture (0.06 g/mL). Then drop the mixture onto the surface of the film (0.25 mL). Allow the ethanol to evaporate before performing the friction test.

2.2 Tribological testing

All tribological tests were conducted on a rotating friction tester (MS-T3001). The rotational speed can be adjusted between 10 to 3000 rpm, and the frictional load can be adjusted between 0 to 20 N. Vacuum friction tests were conducted using the equipment's integrated vacuum chamber, with a relative pressure of -0.9 Pa. All the friction tests conducted in ambient is at room temperature (~ 25 °C) and relative humidity of 20%. The friction experiment used Al_2O_3 balls as the friction pair. Al_2O_3 balls, size 5 mm diameter, precision G10. The Al_2O_3 balls were first cleaned with ethanol for 20 minutes to ensure a clean surface before being mounted on the friction machine. Before the friction test, the surface of the ball was dried in the atmospheric environment to avoid the influence of ethanol on the friction coefficient.

2.3 Materials characterization

The tribofilms were analyzed using HRTEM, EELS (2.0 mm entrance aperture, 0.3 eV channel step size), HAADF, and EDS mapping with a spherical aberration-corrected TEM (JEOL JEM-ARM200F, 200 kV). The corresponding slice samples were obtained via FIB technique (Helios G4 UX, 200 kV). Surface morphology was examined with an SEM (S-4800). The Micro-Raman analysis employed a Raman spectrometer (Micro-Raman Renishaw-invia, 532 nm wavelength), while elemental analysis was done using an XPS (XPS Axis Supra). Crystal structure was determined using an XRD (XRD SmartLab 9KW).

3 Results and Discussions

3.1 Macroscale superlubricity strategy

To further enhance the lubricating performance of the MoS_2 film and even achieve superlubricity, a large number of nanosphere structures are introduced to convert the macroscopic single-asperity contact into a microscopic multi-asperity contact (**Fig. 1**), and enable homogeneous or heterogeneous

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shear at each microscopic multi-asperity contact point. This strategy achieves superlubricity mainly by regulating the topological morphology of materials and constructing heterogeneous structures. It involves the formation of a new friction interface composed of abundant microspheres, which self-assemble into microstructures and convert macroscopic surface contact into microscopic point contact. Furthermore, structural superlubricity is induced at each microscopic point contact. The collective lubrication effect of numerous contact points ultimately realizes macroscale superlubricity.

Based on previous research [24], carbon-plated nanospheres and MoS₂ films are selected to achieve superlubricity. During the friction process, nanospheres are introduced as the core, with MoS₂ wrapped around them, and the reduction of the friction coefficient is achieved through the synergistic action of HFLC and MoS₂. The purpose of introducing nanospheres is to utilize numerous microspheres to transform macroscopic planar contact into microscopic point contact, thereby reducing the contact area. Based on studies of fullerene-like carbon (FLC), its low friction behavior is well-documented. FLC films have a unique nanostructure that forms spherical or graphite shell-like nanoparticles in response to friction, which helps minimize friction and therefore significantly reduces friction [24]. In addition, the hydrogenated carbon films generally have better tribological properties in the open environment. Additionally, there is a possibility that hydrogenated fullerene-like carbon (HFLC) may form heterojunctions with MoS₂. Studies have found that changing the topological morphology of the material itself and actively constructing cage-like structures can reduce the edge effect [25]; there are also related studies observing superlubricity in heterojunctions. Because the lattice parameters at the heterojunction interface are easier to form incommensurate contacts. Compared with the same-phase interface, the heterojunction, which is not affected by the rotation angle of the contact body, has new potential in micro-and macro-scale applications [26]. The combination of HFLC and MoS₂ at the sliding interface further enhances superlubricity by reducing contact area and incommensurate contact [17]. Specifically, we employ SiO₂ nanospheres coated with the HFLC film in conjunction with a MoS₂ film. The hydrogenated fullerene-like carbon coated nanospheres (HFLC

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nanospheres) in friction grasped the MoS₂ sheet to form new nanospheres. When friction occurs, the HFLC/MoS₂ nanospheres form a new structure, which effectively reduces the coefficient of friction (Fig. 1).

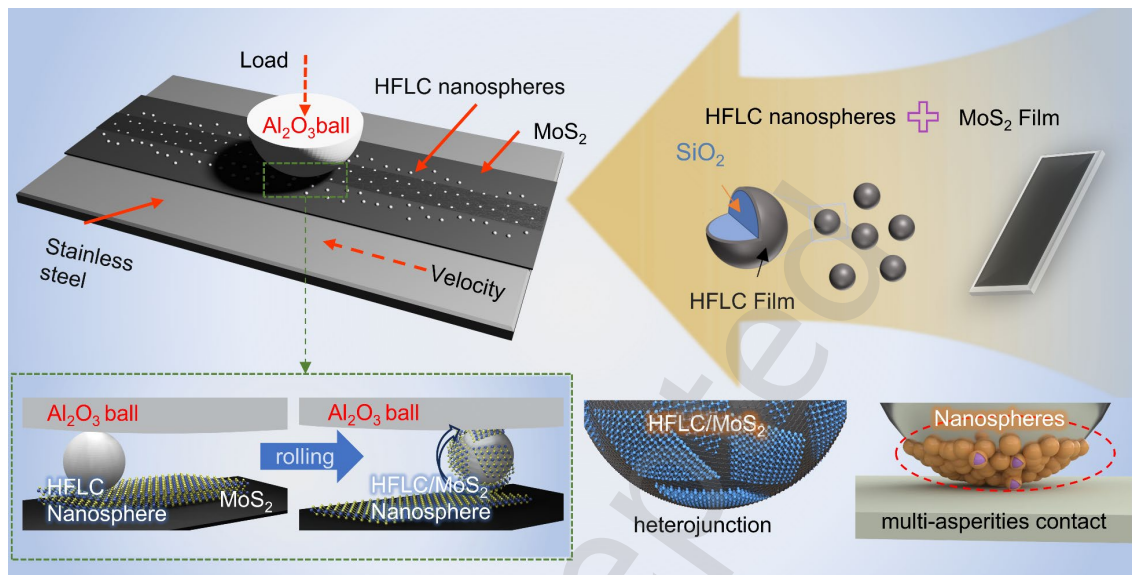


Fig. 1 Macroscale superlubricity strategy by extension of microscale structural superlubricity. Combining HFLC nanospheres with the film forms new lubrication structures during friction, and the tribofilm is composed of many new lubrication structures. Overall, the tribofilm achieves multi-asperity contact and forms a heterojunction at each contact point to ensure lubrication at each point.

3.2 Characterization of the as-prepared MoS₂ films

The weak "van der Waals" interlayer forces of MoS₂ contribute to its low friction coefficient, with the lubrication effect being sensitive to the atmosphere. Atoms located at the edges are more likely to move due to lower coordination and are captured in the smallest energy state of the substrate, leading to edge pinning. Hence, MoS₂ films are prone to failure in O₂ and H₂O-rich environments [27, 28]. For MoS₂ films, the edge effect can be reduced by layer orientation.

The PVD technique was employed to prepare crystalline MoS₂ films (**Fig. 2a**). By adjusting the parameters, the crystallinity of the film was carefully controlled. This enhances the lubrication properties of the film. In comparison to conventional worm-shaped MoS₂ films (Fig. S1). The crystal

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film prepared in this study is smoother and denser than the amorphous film (Fig. 2b and Fig. S1). The XRD spectra results show that the MoS₂ nanoflakes in the film are mainly oriented along the (002) plane (Fig. 2c). This result indicates that the film has a crystal structure with layered order. To gain further insights into its structural composition, Raman spectroscopy analysis was conducted on the MoS₂ film (Fig. 2d). The vibrational peaks at 379 cm⁻¹ and 407 cm⁻¹ were assigned to E_{2g}¹ and A_{1g} modes of MoS₂, respectively [29]. Additionally, XPS was utilized for investigating element valence states in this film (Figs. 2e–g). Notably, apart from observing a smaller shoulder peak near 226 eV in the Mo 3d spectrum attributed to S 2s peak presence, another small shoulder peak (235 eV) corresponding to molybdenum oxide species was also detected. This observation suggests surface oxidation of molybdenum species present in the film. Comprehensive characterizations confirm that layered MoS₂ films with preferential (002) crystal plane orientation are successfully deposited on the stainless steel substrate. Furthermore, compared to amorphous MoS₂, the enhanced ordering along (002) planes are associated with a lower friction coefficient. In a vacuum, the friction coefficient of amorphous MoS₂ film is measured to be 0.25, whereas for MoS₂ films with a certain degree of crystallinity, the friction coefficient decreases significantly to only 0.04 (Fig. S2). When exposed to open-air, MoS₂ films with a specific level of crystallinity exhibit a reduced friction coefficient of approximately 0.06 (Fig. S2). Conversely, the amorphous MoS₂ film demonstrates an elevated friction coefficient in an open environment, reaching as high as 0.7 (Fig. S2). These films effectively exploit the layered molecular structure of MoS₂ to take advantage of its weak van der Waals force characteristics, thereby resulting in favorable lubricating properties under specific atmospheric conditions. However, the issue of incomplete elimination of the "edge effect" persists in these pure MoS₂ films, resulting in disparate friction coefficients and wear life between vacuum and open atmospheric environments. Our strategy with the MoS₂ and HFLC combination fully leverages the weak van der Waals forces between the layers of the 2D materials, forming heterojunctions that reduce the sensitivity of the lubrication structure to atmospheric conditions.

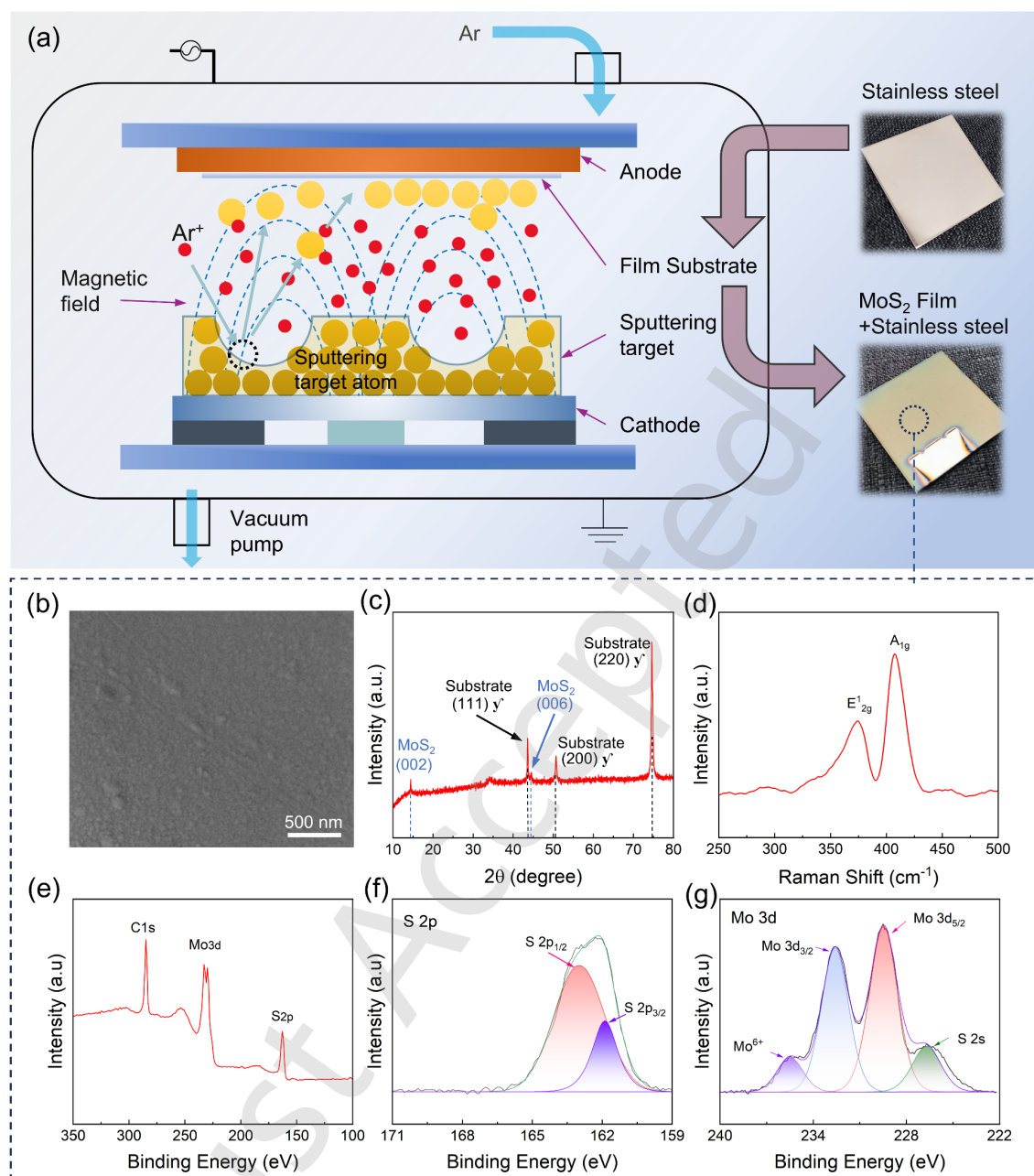


Fig. 2 Preparation and characterization of MoS₂ film. a) Schematic representation of the sputtering process for film deposition. b) SEM of the MoS₂ film. c) XRD patterns of the MoS₂ film. d) Raman spectroscopy of the MoS₂ film. e–g) XPS spectrum of the MoS₂ film (e), and S 2p (f) and Mo 3d (g) spectra.

3.3 Characterization of the as-prepared HFLC nanospheres

The preparation of hydrogenated fullerene-like carbon coated nanospheres (HFLC nanospheres) using

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PECVD technology (**Fig. 3**). Ion assistance during the chemical vapor deposition (CVD) process enhances crosslinking of the films and promotes the formation of fullerene-like (FL) structures. The deposition parameters, including ion energy and flux, can be adjusted to control this process. The formation mechanism of FL microstructures in amorphous carbon films, as detailed in previous researches, involves several key processes [24, 30, 31]. These include the specific deposition conditions of DC-PECVD, the critical role of hydrogen in stabilizing sp^2 hybridized carbon atoms, and the microstructural evolution facilitated by hydrogen incorporation. Intrinsic stresses and strains within the film foster curved and cross-linked carbon networks, reducing surface energy and stabilizing the FL configuration. These mechanisms endow amorphous carbon films with distinctive mechanical and tribological properties.

The surfaces of all HFLC nanospheres appeared darker (Fig. 3a). The SEM images showed that each group of nanospheres had a regular shape without any obvious deformation (Fig. 3b and Fig. S3a). The carbon content of each point on a single nanosphere was determined by EDX elemental analysis, and the highest carbon content was 21.05% (Fig. S3b). The carbon content of other parameters is 18.97%, 16.94%, and 14.09%, respectively (Fig. S3b). Based on the carbon content, the samples prepared using the sample with the highest carbon content were selected for further experiments. Element mapping distribution using EDS scanning demonstrated an even distribution of carbon elements on each ball's surface (Fig. 3b). The carbon content of the scanned area is the same as that of the point scan result, which is about 22% (Fig. 3c). Detection results verify the presence of a carbon film on the nanospheres' surface.

Further tests are required to investigate the crystallographic structure of the carbon film. XPS analysis (Fig. S3c) of the C 1s spectrum identified peaks at approximately 284.0 eV, 285.0 eV, and 288.1 eV, corresponding to sp^2 hybridized bonds, sp^3 hybridized bonds, and C=O bonds, respectively [32]. Conventional EDS and XPS cannot measure the hydrogen element content. By means of elastic recoil detection (ERD) spectroscopy, it is possible to determine whether the carbon film contains

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hydrogen. The experimental results showed the presence of hydrogen in the carbon film (Fig. 3d). XRD is commonly employed for the determination of the crystalline structure of materials. It can be utilized to assess the crystallinity of carbon films. The results before and after coating exhibit a peak cluster only at 20 degrees (Fig. 3e), and a comparison of the two curves reveals no significant alterations. These patterns are in accordance with the characteristic XRD profiles of amorphous SiO₂ materials [33]. No characteristic carbon related peaks were detected in the XRD analysis results following the coating process (Fig. 3e). However, the color and elemental composition of the powder indicate the presence of carbon. Therefore, it can be inferred that a non-crystalline amorphous carbon film covered the surface of SiO₂ nanospheres after PECVD treatment.

To further validate this inference, TEM observations of the carbon film were performed (Fig. 3f). Upon closer inspection, the carbon film was found to have an overall amorphous structure with many fullerene-like structures. In a non-crystalline film, numerous single-layer graphene sheets can be observed twisted in random orientations (Fig. 3f). The corresponding selected area electron diffraction (SAED) pattern displays three diffuse rings with spacings of approximately ~ 3.4 Å, ~ 2.0 Å, and ~ 1.2 Å from the inside out. The latter two spacings are similar to those of amorphous carbon, while the ~ 3.4 Å spacing matches that of layered graphite. This twisting phenomenon can be attributed to the formation of not only the hexagonal carbon rings during the coating process, but also carbon rings with odd numbers of sides, such as pentagons or heptagons. Odd ring formations cause the graphene basal planes to curve [34]. A shoulder peak at 1200 cm^{-1} was detected by Raman spectroscopy (Fig. 3g). This peak is usually attributed to a five- or seven-sided ring of hexagonal carbon atoms [35]. Further analysis of the peaks revealed the presence of the $7A_1$ peak at 1200 cm^{-1} and the $5A_1$ peak at 1470 cm^{-1} (Fig. 3g). It is noted that the peaks at 1200 cm^{-1} and 1470 cm^{-1} are attributed to pentagonal and heptagonal structures, respectively [36]. The Raman test results are coincident with the TEM images, further confirming that the carbon film covering the surface of the nanosphere powder is mainly composed of amorphous non-crystalline carbon (a-C:H). The carbon film contains graphene layers

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with a fullerene-like structure. Thus, the SiO₂ nanospheres are coated with an HFLC film after PECVD treatment.

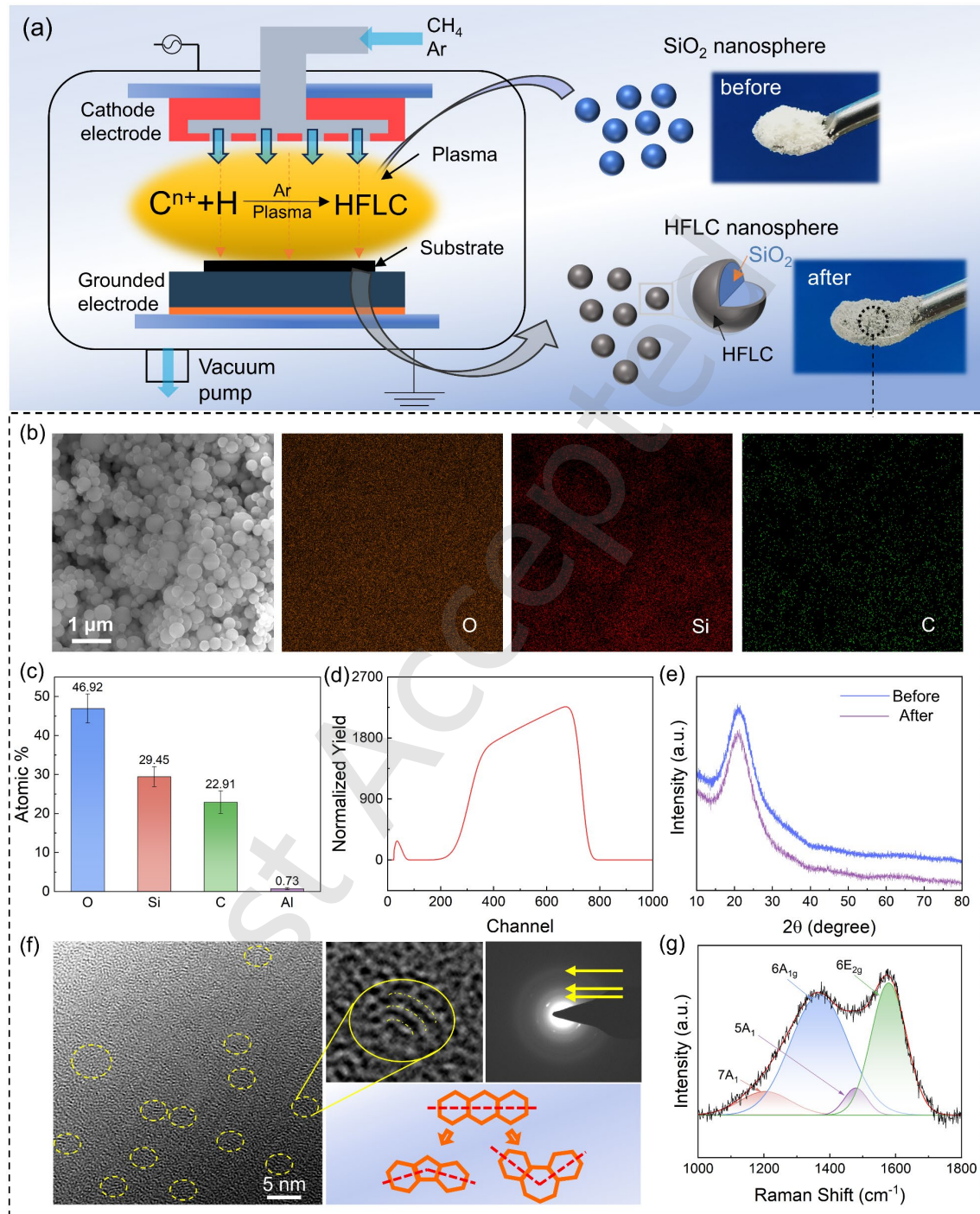


Fig. 3 Fabrication and characterization of the HFLC nanospheres. a) Schematic diagram of the fabrication process of HFLC nanospheres. b) Surface morphology of HFLC nanospheres and elemental distribution of O, Si and C. c) The proportion of O, Si, and C from (b). d) Hydrogen signals obtained by ERD from the HFLC nanospheres. e) XRD of the HFLC nanospheres. f) TEM images of HFLC

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nanospheres and their electron selective diffraction patterns. g) Raman spectra of the HFLC nanospheres.

3.4 Superlubricity behaviors

As mentioned previously, the low friction behavior of pure MoS₂ films is sensitive to environmental conditions. The aim of this strategy is to form lubricated structures that are insensitive to environmental conditions, so HFLC/MoS₂ nanospheres were designed. To demonstrate structural stability, friction behavior tests were conducted on MoS₂ and HFLC/MoS₂ under different atmospheres.

We combined MoS₂ films with HFLC nanospheres and performed friction experiments (**Fig. 4**). Tests were conducted in both vacuum and open atmospheric environments (Figs. 4a–f and Fig. S4). With the addition of HFLC nanospheres, the friction coefficient under vacuum conditions was 0.005. In an open environment, the friction coefficient reached 0.009 (curves 4 and 6). The pure MoS₂ films used in this experiment had a friction coefficient of 0.05 under vacuum conditions and 0.07 in open air (curves 2 and 3). The addition of HFLC nanospheres reduced the friction coefficient from 0.05 to 0.005 in vacuum and from 0.07 to 0.009 in open air. The difference in friction coefficients of pure MoS₂ films between vacuum and air originates from that oxygen and water molecules in air react with MoS₂ during friction to form MoO₃, which destroys the layered lubrication structure of MoS₂ [37]. As for the difference in friction coefficient between vacuum and air after adding HFLC nanospheres, in addition to the environmental sensitivity of MoS₂, our previous studies have shown that water molecules can adsorb on the surface of hydrogenated amorphous carbon and inhibit graphitization. Meanwhile, water molecules generate capillary force to increase the interfacial friction resistance. Furthermore, water molecules participate in tribo-oxidation, impair the lubrication performance, and offset the passivation effect of hydrogen [38]. However, under the same environmental conditions, the incorporation of HFLC microspheres exhibits a prominent and excellent friction-reducing effect.

Additionally, the film's lifespan was extended in an open atmosphere. The addition of pure SiO₂

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nanospheres did not form an effective lubricating structure, with a friction lifespan of less than 2500 cycles and no stable friction coefficient was observed (curve 1). To further verify that the combination of MoS₂ films and HFLC nanospheres can form a heterostructure with low atmospheric sensitivity, we conducted variable atmosphere experiments (curve 5). The friction coefficient remained at 0.005 during 0 ~ 10000 cycles in a vacuum environment. At the 10000th cycle, the atmosphere was changed to an open environment, and no significant change was observed (remaining at 0.009). These experiments demonstrate that the combination of MoS₂ films and HFLC nanospheres effectively reduces the friction coefficient and forms a lubricating structure with low atmospheric sensitivity. It was noted that in curve 4, it took approximately 3700 sliding cycles to stabilize into the superlubricity state, whereas the running-in periods of curves 5 and 6 were significantly shorter than that of curve 4. A probable reason is that for nano-particles (NPs) strongly embedding into the solid film matrix, the run-in period is long. Because the NPs' releasing and nanoscrolls' formation need more time. However, for the condition similar to this work. However, when the NPs are externally added into the friction interface, the run-in time is shortened.

Microscopic images of the wear tracks and element distribution can also reflect the lubricating effects of the introduced HFLC nanospheres. By comparing the distribution of Mo, S, and C elements on the wear tracks (Figs. 4b–f), it can be considered that a new structure is formed. Observing the friction results after adding microspheres (Figs. 4b–d), it can be observed that Mo, S, Si, and C elements are distributed on the wear tracks. This indicates that the HFLC nanospheres participate in the friction process and form a new lubricating structure.

By comparing the results of the friction experiments under different atmospheric conditions, the wear track morphology and element distribution in the variable atmosphere experiment (Fig. 4c) are coincident with those under constant atmosphere experiment (Figs. 4b, d and Fig. S4). This indicates that the newly formed lubricating structure has low atmospheric sensitivity. In contrast, the addition of SiO₂ alone does not provide a lubricating effect, as no Mo, S, or Si elements are found on the wear

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tracks (Fig. 4e). This element distribution indicates that the MoS₂ film has been completely consumed and is not able to form an effective lubricating structure. The wear scars of pure MoS₂ films in an open atmospheric environment show surface morphology and element distribution like those with only SiO₂ added (Figs. 4e, f). This demonstrates that pure MoS₂ films struggle to form an effective lubricating structure in an open atmosphere. By comparing the wear tracks (Figs. 4b–f), it can be confirmed that the HFLC nanospheres participate in the friction process, forming a lubricating structure with low atmospheric sensitivity.

Based on the designed lubrication strategy, the formation and accumulation of HFLC/MoS₂ nanospheres are the main reasons for the occurrence of superlubricity. The introduction of HFLC nanospheres during the friction process leads to the folding of MoS₂ layers. By encapsulating MoS₂ on the exterior of the spheres to form HFLC/MoS₂ nanospheres, the exposed edge dangling bonds can be reduced, which effectively lowers the coefficient of friction. After the formation of the HFLC/MoS₂ nanosphere structure, the outer HFLC/MoS₂ layer can be utilized as a heterojunction. This 2D heterojunction with weak interlayer van der Waals interactions and inherent lattice mismatch properties is an excellent platform for achieving ultra-low friction independent of twist angle [18, 26, 39]. The HFLC/MoS₂ heterojunction can also play a role in passivating edge dangling bonds, thus achieving long-lasting lubrication in air [39]. After the formation of HFLC/MoS₂ nanospheres with regular spherical structures, they can function as nano-bearings. The aggregation of numerous HFLC/MoS₂ nanospheres forms a new tribological layer, in which the contact state transforms from macroscopic surface contact to microscopic multi-point contact due to the presence of numerous spherical particles, thereby reducing the real contact area. Moreover, owing to the encapsulation by MoS₂, the multiple shear interfaces include homogeneous and heterogeneous shear interfaces such as MoS₂/MoS₂, HFLC/HFLC, and MoS₂/HFLC. The reduced real contact area and diverse interfaces prone to shear sliding act synergistically, thus achieving a superlubricity state.

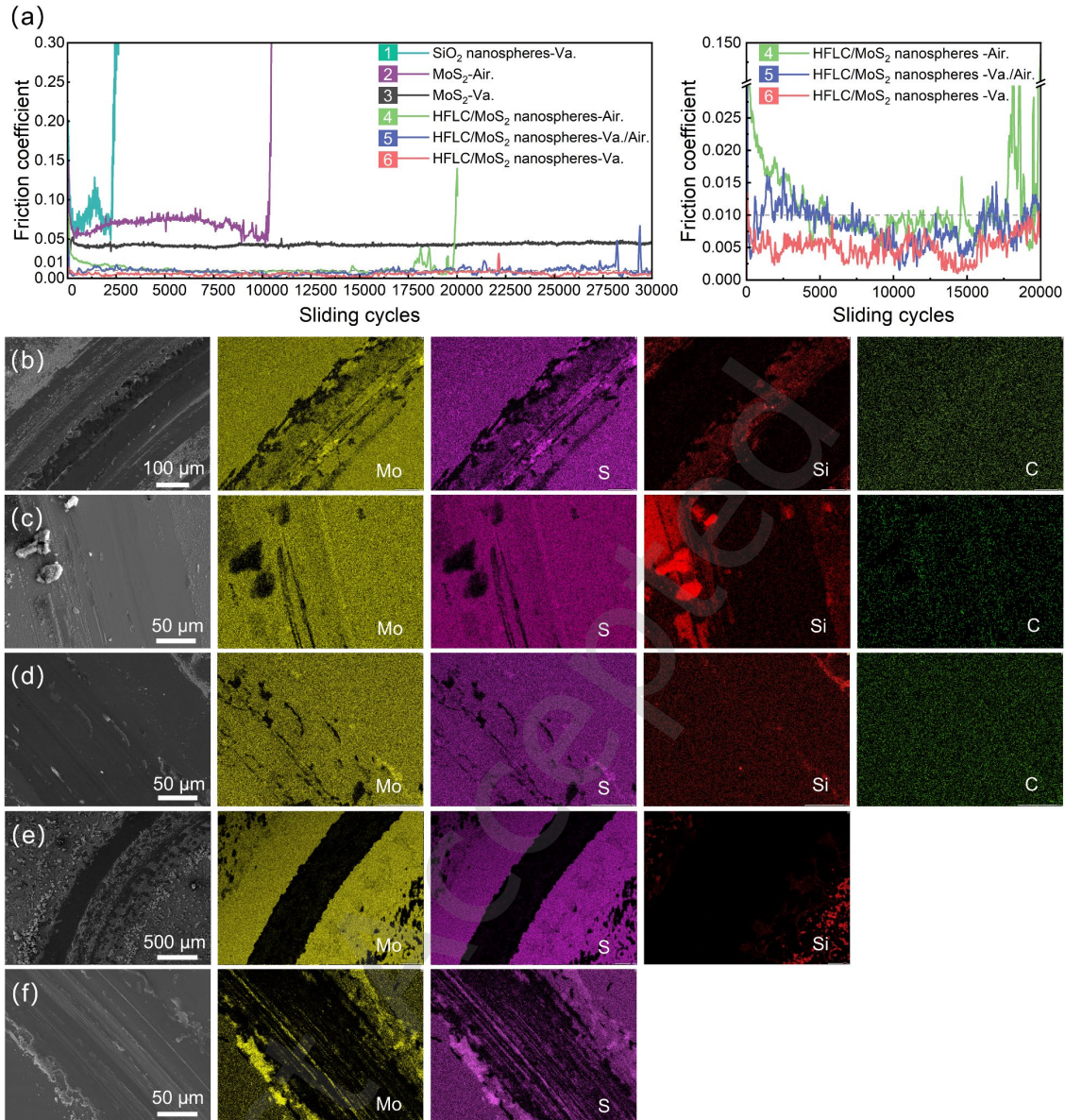


Fig. 4 Friction behavior and wear track characteristics. a) Friction coefficient curves under different friction conditions (left) and a magnified view of the friction coefficient curves in the superlubricity state (right). b–d) SEM and EDS-mapping of wear tracks from (a): (b) atmospheric condition (curve 4); (c) variable atmosphere condition (curve 5); (d) vacuum condition (curve 6). e) Wear track with SiO₂ nanospheres in vacuum (curve 1). f) Wear track with pure MoS₂ in ambient (curve 2).

Similar theories and lubrication strategies have been proven to be feasible and effective in other researchers' work [22, 36]. The heterojunction has been proven to exhibit excellent lubrication

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properties [18, 26]. Macroscopic superlubricity has more limitations compared to microscopic superlubricity. Nanodiamonds can act as nanoball bearings when used as additives and coated with graphene, providing additional mechanical strength [22, 40]. For example, it has been observed that the friction coefficient between MGNFs/HOPG (multilayer graphene nanoplatelets/highly oriented pyrolytic graphite) is two orders of magnitude higher at the macroscopic scale than at the nanoscale, indicating that there are additional frictional energy dissipation channels at macroscopic scale [20]. Studies have found that the in-situ formation of OLC (onion-like carbon structures) on the sliding interface can reduce the contact area and form incommensurate contact with the OLC surface, thereby successfully demonstrating superlubricity [41]. Researchers have specially designed a three-dimensional egg-carton structure to achieve the transition from macroscopic planar contact to microscopic point contact, thereby realizing excellent lubrication effects [13].

In this study, the introduction of abundant solid nanospheres enables the formation of a continuous protruding contact surface like a cobblestone road surface. These continuous protrusions transition the macroscopic planar contact to microscopic point contact, thereby reducing the limitations of macroscopic superlubricity. Based on the friction experiment results and the analysis of the HFLC/MoS₂ nanosphere structure designed in our strategy, the tribological layer formed by the accumulation of numerous HFLC/MoS₂ nanospheres is the core of superlubricity. The presence of this new tribological layer transitions macroscopic planar contact to microscopic point contact, each of microscopic point contains a HFLC/MoS₂ layer. This tribological layer can control interface order, weaken chemical interactions, and manage mismatched configurations, thus meeting the prerequisites for structural superlubricity. Additionally, the HFLC/MoS₂ nanospheres can serve as an excellent platform, enabling various homogeneous and heterogeneous shear interfaces formed during friction to function effectively. We attribute the achievement of the superlubricity state to the synergistic effect of the aforementioned friction reduction pathways.

To validate our hypothesis and investigate the specific mechanisms behind superlubricity, the

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factors influencing the friction coefficient can typically be derived from two surfaces and one interface, namely the wear scar surface, the wear track surface, and the interfacial tribofilm. By analyzing the morphology of the wear scars, they were considered that the presence of HFLC nanospheres and MoS₂ (**Fig. 5**). Optical microscopy analysis showed that the wear scars were predominantly a mixture of brown and gray-white colors (Fig. S5). The varying shades of brown indicate the varying proportions of gray-white and brown substances. By comparing our wear scar sample with other wear scar samples (Fig. 5a and Fig. S5). We infer that the wear scar consists of a mixture of MoS₂ and the coated nanosphere. To validate this hypothesis, the wear scars were examined using SEM and subsequently analyzed via energy dispersive EDS. The results show (Fig. 5a) that the wear scar samples contain Mo, S, O, Si and C elements. These elements are consistent with the MoS₂ and the coating nanospheres. The SEM technique can only provide morphological information and lacks color information. Nevertheless, by integrating the optical microscopy images and elemental distribution data, it was possible to identify areas of higher molybdenum and sulfur content corresponding to the darker brown areas observed under optical microscopy. Silicon and carbon were widely distributed in the sample, partially overlapping with the brown areas described above. Accordingly, the brown substance detected after friction is MoS₂, while the white component corresponds to HFLC nanospheres. The wear scar presents a complex structure composed of these two substances.

By magnifying the local morphology of the wear scars (Figs. 5b–d), the mixed state of MoS₂ and HFLC nanospheres can be better understood. This facilitates the analysis of the mechanisms underlying superlubricity. In the MoS₂-rich region (Fig. 5b), the surface tends to be smooth. However, upon closer examination, it is still possible to observe spherical-like substances embedded within the surface (spot line in Fig. 5b). From the element distribution map, it can be observed that Mo, S, O, Si, and C elements are mixed throughout the entire area with no obvious zoning. Only the Mo and S signals are stronger in the smooth part. However, there are still a host of HFLC nanospheres in the MoS₂-rich region. Small spherical particles were observed in areas where the content of MoS₂ was

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relatively low (Fig. 5c). Based on the corresponding elemental distribution map, these small spherical particles were identified as HFLC nanospheres. This region also contains MoS₂. By conducting a comprehensive analysis of the elemental distribution in the wear scar, and combining the morphological features observed in each region, we propose that the entire wear scar is formed by a combination of HFLC nanospheres and MoS₂. The wear scar on the surface of the spherical structure exhibits prominent protrusions and depressions, characterized by numerous densely packed undulating waves (spot line in Fig. 5c). In summary, the wear scar surface exhibits a pattern consisting of countless protrusions, characterized by "grain undulations and grain texture". When associated with actual objects, this morphology can be referred to as "candy-like texture" (like the texture produced by a pile of beans or marbles in a bowl).

The inside of a wear scar can be further understood by looking at the fracture of a wear scar (Fig. 5d). The surface state of the wear scar at this position is similar to that of Figs. 5b, c, with a mixed distribution of Mo, S, O, Si, and C (Fig. 5d). This is consistent with the element distribution results in Fig. S5. On the fracture surface, the small spherical morphology within the etch pits was observed to be regularly layered, and the small balls were found to be wrapped in certain substances (Fig. 5d). Furthermore, the small spheres were wrapped in certain substances. Based on the element distribution results, we concluded that these spherical objects were HFLC nanospheres, with MoS₂ adhered to their surfaces. Observation of this phenomenon can further confirm the existence of the HFLC/MoS₂ Nanosphere. A large quantity of nanospheres aggregate to generate a fresh tribofilm, whereby the wear scar surface displays dense continuous protrusions and a granular microstructure (Figs. 5c, d). The observed phenomena support our hypothesis that the HFLC/MoS₂ nanospheres act as a tribofilm, effectively reducing the contact area.

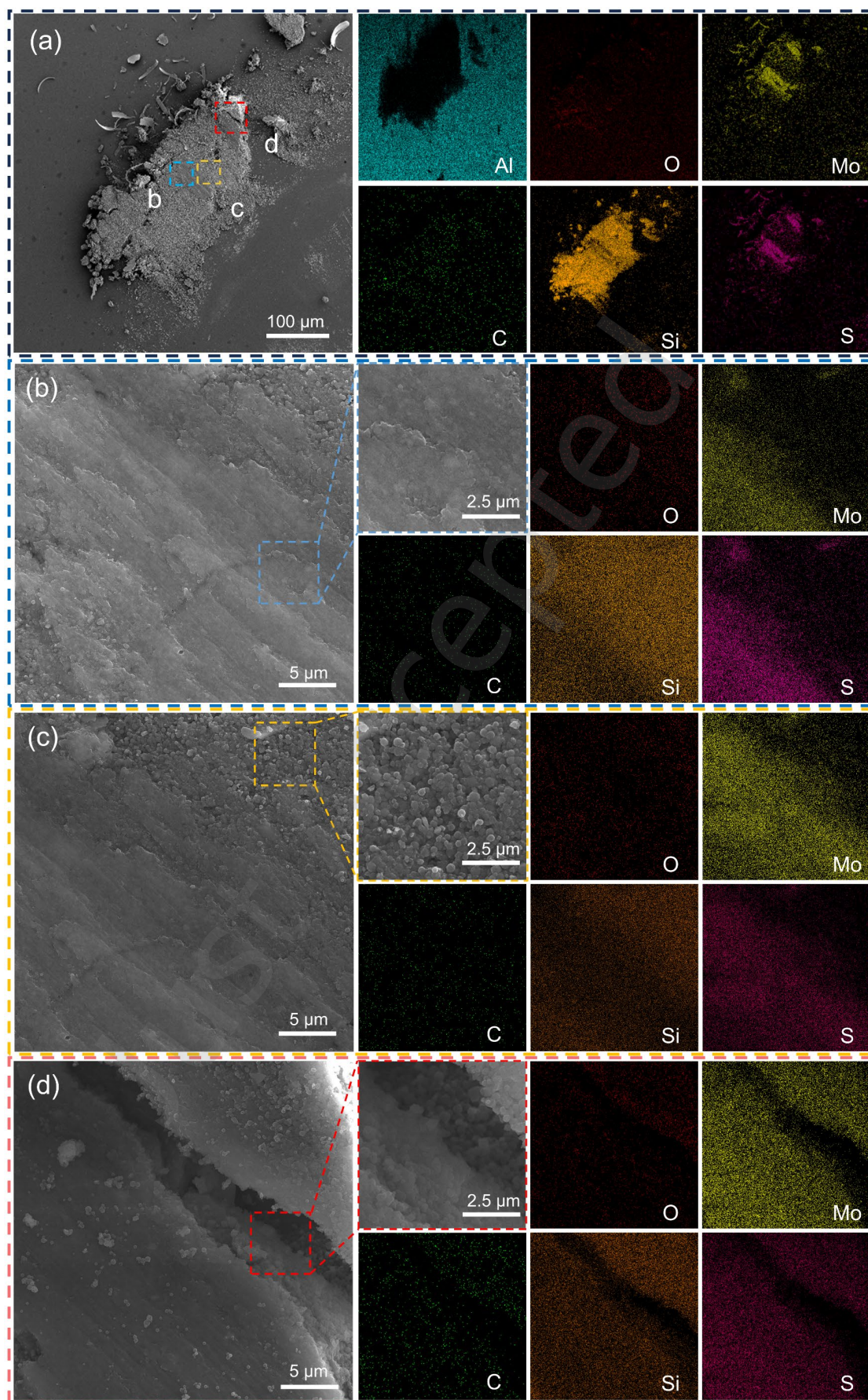


Fig. 5 SEM and EDS-Mapping images of the wear scar (curve 5 in Fig. 4). a) Morphology and

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elemental distribution of wear scars formed under variable atmosphere conditions. b–d) Magnified view of the corresponding area in (a). Surface morphology of wear scars and corresponding elemental distribution (b, c). d) Morphology of the fracture area and its corresponding elemental distribution.

Further, HFLC/MoS₂ nanosphere structures were found on the wear track (**Fig. 6**). The corresponding elemental distribution map (Fig. 6a) reveals an even distribution of Mo and S signals. Considering the associated wear track morphology (Figs. 6a, b), it can be inferred that the MoS₂ film was not eliminated. The area is signalized by silicon, oxygen and carbon, with carbon covered microspherical bodies on the outside of the wear track. However, no obvious spherical substances were found in the interior (Figs. 6a, b and Fig. S6). The entire wear track region maintains a smooth surface. Si is enriched in the friction trace and appears as an accumulation of particulate matter (Figs. 6a, b). This phenomenon is also confirmed in Fig. 6 and Fig. S6. Further, enlarge the accumulation area (square area), the area is similar to the accumulation state of nanospheres on the wear scar, and the shape of the small balls can be seen on the surface. It can be concluded that during the friction process, the nanospheres mix with the peeled-off MoS₂ and then form a new tribofilm, which changes the contact mode.

XRD and Raman spectra were used to analyze the wear track phase structure. The Raman spectra showed only carbon related D and G peaks, consistent with pre-friction powder test results (Fig. 6c). XRD spectra revealed a prominent peak at 14.1° for the 002 reflection of MoS₂, along with substrate material peaks (Fig. 6d). However, the 002 peak of MoS₂ appeared in a broader peak form than the original film. After combining the XRD spectra of the nanospheres, it was found that the peak package at 20° corresponds to the amorphous SiO₂. The wear tests yielded similar results to the XRD peaks of the nanospheres, and the positions of the two substances were similar, resulting in the effect shown in the Fig. 6d. This is consistent with previous results on amorphous carbon films test as XRD showed the presence of MoS₂ and HFLC nanospheres on the wear track and no obvious C-related peaks were

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observed. Based on the above test results, morphology images, and element distribution maps, it can be inferred that a new tribofilm is formed by mixing the nanospheres with MoS₂ during the friction process.

The morphology observed from the wear scars and wear tracks (Fig. 5 and Fig. 6) matches well with the morphology of the tribofilm formed by the envisaged HFLC/MoS₂ nanospheres. This tribofilm, composed of numerous microspheres, divides macroscopic contact into nanoscale contacts, reducing the actual contact area compared to pure MoS₂ film friction experiments. The introduction of HFLC nanospheres results in the accumulation of a layer of nanospheres accumulating on the surface of the grinding ball. The nanosphere accumulation forms a surface pattern similar to a cobblestone paved road (as illustrated in the schematic in Fig. 1 and Fig. 5d). The entire surface can be referred to as "candy grain texture", a surface composed of countless raised points arranged in a regular pattern. The large surface dominated by these points reduce the contact area compared to the original alumina abrasive balls. Additionally, research has shown that the macroscopic realization of superlubricity is more limited than at the microscopic level, with other friction energy dissipation channels existing at the macroscopic scale. This includes structural defects in the preparation of the material, which suppress superlubricity when the contact area is large [20]. Structural defects at the molecular level are difficult to avoid in actual manufacturing. Macroscopic large surfaces are composed of countless microscopic small structures. It is easier to eliminate defects and achieve uniform orientation at single points than on the entire macroscopic surface. By dividing the macroscopic scale into countless small subdivisions, it is easier to achieve uniform orientation and to form a defect-free state in the smaller parts. This can reduce the macroscopic defects that affect the coefficient of friction. This design has been supported by relevant literature in achieving macro-superlubricity performance [20, 23]. Surface observation reveals that nanospheres take part in friction and assemble into a unique structure constituting the new tribofilm.

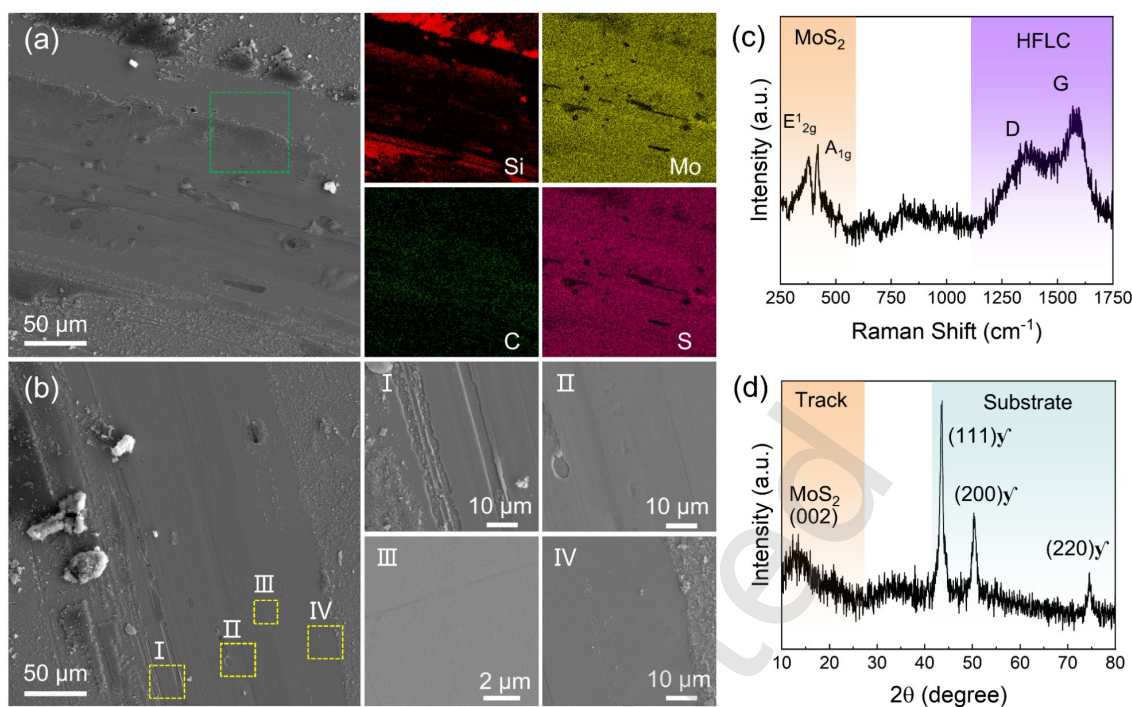


Fig. 6 SEM and EDS-Mapping images of the wear track (curve 5 in Fig. 4). a, b) Morphology and elemental distribution of wear tracks formed under variable atmosphere conditions. c, d) Raman spectra (c) and XRD spectra (d) of the wear track centers.

3.5 HFLC/MoS₂ nanospheres self-assembling mechanisms

To further clarify the composition of the tribofilm, the mechanism of MoS₂-encapsulated microspheres, and their correlation with the superlubricity performance. The nanostructures of the tribofilm were resolved at the atomic scale based on morphology and elemental distributions (Fig. 7 and Figs. S7–11). The top of the slice was protected by platinum for FIB processing, and the bottom was an alumina grinding ball, with the tribofilm being the middle part. The entire tribofilm can be represented by this small section (Figs. 7a–c). The structure is composed of small spheres, with regular morphology within the tribofilm (Fig. 7a). Preliminary observations show that the tribofilm is made up of abundant well-defined spherical particles, mostly with a diameter of 500 nm. This indicates that the tribofilm is formed by the accumulation of numerous nanospheres. Therefore, it can be hypothesized that these nanospheres act as nano-bearings to attach and support the lubricating phase. The shape of the small

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spheres near the wear track surface (those near the Pt layer) are also regular. The top layer of spheres forms a continuous undulating protrusion on the side closest to the pitting surface. Some spheres are covered with Pt, but their outlines can still be observed. This phenomenon specifically explains the formation of the "sugar granule texture" on the wear scar surface (Fig. 5). Overall, the tribofilm is formed by the accumulation of numerous spheres, consistent with the previously observed morphology of the wear scar surface. In magnified regions, the spherical morphology remains regular, with other materials attached to the edges (Fig. 7b and Figs. S7–11). The entire small sphere is composed of Si and O elements, while the attached substances at the edges are composed of C, Mo, and S elements (Fig. 7c). This phenomenon is consistent with the conclusion that the HFLC nanosphere surface is wrapped by MoS₂. Thus, it can be preliminarily concluded that HFLC/MoS₂ nanospheres are formed and the entire tribofilm is composed of HFLC/MoS₂ nanospheres.

Analysis of TEM images confirms that macroscopic superlubricity can be achieved through a tribofilm composed of HFLC/MoS₂ nanosphere structures. A single HFLC/MoS₂ nanosphere structure is formed by carbon-plated nanospheres encapsulating MoS₂. The HFLC/MoS₂ structure on the outermost layer of the microspheres can be considered as a heterojunction. When these two different types of 2D materials are stacked together, they exhibit the electronic properties of covalent/ionic heterojunctions. This effectively blocks strong chemical interactions between adjacent covalent compounds and facilitates structural superlubricity [13, 42]. Covalent compounds (such as graphene and boron nitride) have delocalized π bonds with directionality and saturation. However, MoS₂ is an ionic compound that lacks directionality and saturation and produces a screening effect [43, 44]. Therefore, the friction coefficient can be reduced by combining carbon-plated nanospheres with MoS₂ films to form a HFLC/MoS₂ layer. This ensures lubrication at the nanoscale contact points on individual nanospheres. The tribofilm is composed of numerous HFLC/MoS₂ nanospheres stacked together. Although each sphere is small under the microscope, when these small spheres accumulate at the friction interface, the superlubricity at numerous points results in robust macroscopic

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superlubricity [43, 45]. The accumulation of small spheres forms a tribofilm that alters the contact mode. Compared to the lubrication method of pure MoS₂ films, the presence of numerous nanoscale microspheres transforms the macroscopic contact of pure MoS₂ films into numerous nanoscale contacts. The tribofilm formed by HFLC/MoS₂ nanospheres reduces the actual contact area, lowering the friction coefficient. The formation of the HFLC/MoS₂ nanosphere tribofilm converts the original single homogeneous MoS₂/MoS₂ shear interface into a combination of coexisting MoS₂/MoS₂, HFLC/HFLC and MoS₂/HFLC shear interfaces. The tribofilm contains many spherical structures. The introduced HFLC nanospheres and the HFLC/MoS₂ nanospheres formed during the friction process all remain intact spherical shapes, demonstrating that this multi-point contact platform has favorable load-bearing capacity. Based on the above analysis, this lubrication mechanism integrates various friction-reduction approaches. Their cumulative synergistic effect lowers the friction coefficient and enables macroscopic superlubricity.

To further understand how HFLC and MoS₂ function under the action of nanospheres, the tribological layer's spheres were observed at a higher magnification (Figs. 7d–h). Further magnification reveals the appearance of nanocrystalline graphite (NG) [24] on the surface of the spheres. The dark substance attached to the edges of the small balls contained many quantum dot lattices (Figs. 7b–g and Figs. S7–11). In conjunction with Fig. 7b and its corresponding element distribution, these black substances were identified as a mixture of carbon and MoS₂. By measuring the spacing between the quantum dot lattices in the dark area (Fig. 7f), the inter-lattice spacing was found to be 0.22 nm. Combining the element distribution with the identification of carbon quantum dots [46]. Additionally, the experiment found that the structure of carbon quantum dots is identical to the nanocrystalline structure observed in previous studies [24]. Subsequently, carbon quantum dots will be referred to as nanocrystalline graphite (NG).

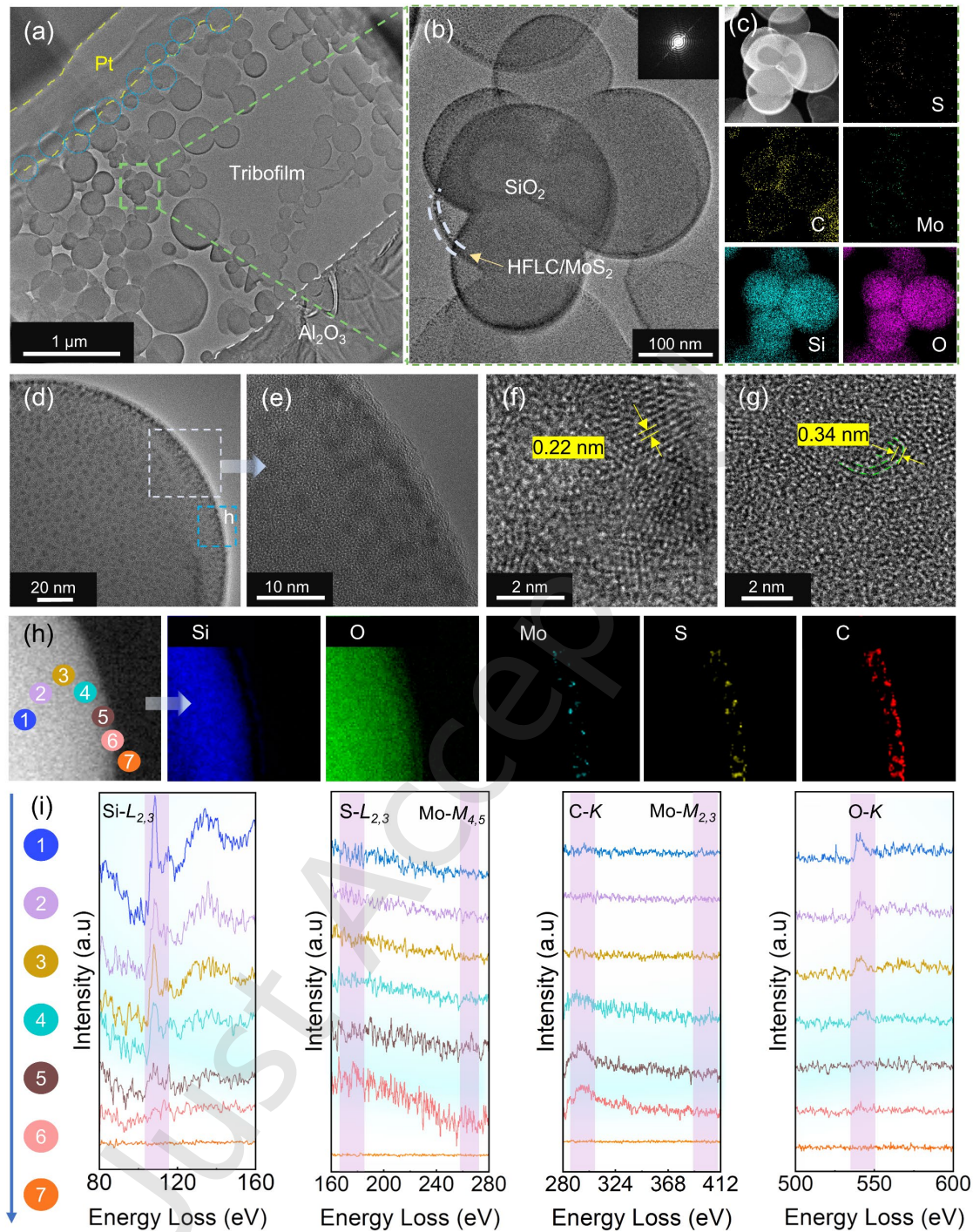


Fig. 7 FIB-STEM-EELS analysis of the tribofilm (curve 5 in Fig. 4). a) Overall TEM image of the tribofilm (The tribofilm was formed under variable atmosphere conditions). b) Magnified image of HFLC/ MoS_2 nanospheres. c) HAADF-STEM images and element distribution in (b). d, e) HRTEM of single nanosphere edge region. f, g) NG structures (0.22 nm) (f) and HFLC structures (0.34 nm) (g) were observed within the single nanosphere edge region. h) Electron energy loss spectroscopy (EELS) mapping in (d). i) EELS at specific points (1–7) on (h).

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A large amount of NG was found on the HFLC nanospheres after friction (Figs. 7b–g and Figs. S7–9). Before the friction, there are a lot of curved graphene sheets with fullerene-like structures on the carbon film. After friction, the NG on the nanospheres is more clearly visible than the initial layered structure of the curved graphene sheet (Fig. 3f, Figs. 7b–g and Figs. S7–11). Also found the FL structure has not completely into the NG structure (Fig. 7g). During the friction process, disordered amorphous carbon films and curved graphene sheets with fullerene-like structures are transformed into layered, ordered graphene sheets. The disordered amorphous carbon film and the curved graphene sheets within it exhibit a transformation under the influence of friction. The friction process converts disordered graphene layers into layered and ordered graphene sheets. Related studies have shown that disordered amorphous carbon films can be transformed into ordered graphene layers under the action of certain forces and temperatures [34, 41, 47]. Existing studies have demonstrated that HFLC possesses intrinsically curved and highly cross-linked graphene basal planes, which hardly undergo large-scale interlayer sliding. Consequently, only low-molecular-weight nanoparticle-like wear products (e.g., NG) can form at the friction interface. These products effectively shield and eliminate strong interfacial carbon–carbon interactions, greatly reducing the interfacial shear resistance. Furthermore, such wear products can divide the original macroscopic full-surface contact into discrete microscopic point contacts. The actual contact area is remarkably reduced, and the adhesion and shear resistance at the contact interface are substantially weakened, thereby providing structural prerequisites for achieving ultra-low friction and macroscale superlubricity [34].

By analyzing individual small balls (Figs. 7d–h), a more specific understanding of the “HFLC/MoS₂ nanosphere” can be obtained. It is hypothesized that the centers of the individual spheres are SiO₂ and the edges are a mixture of MoS₂ and C. This hypothesis is supported by electron energy loss spectroscopy (EELS) obtained from inside-out pickup sites (Points 1–7 in Fig. 7h) on individual nanospheres, which quantitatively elucidated the “HFLC/MoS₂ nanosphere” structure (Figs. 7h, i). The main characteristic of the S-L_{2,3} is 166.5 eV, while the Mo consists of Mo-M_{4,5} (267 eV) and Mo-M_{2,3}

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(404.5 eV). The Si-L shows a peak at 99.4 eV, and the O-K at 532 eV. The C-K, with characteristic π^* and σ^* features, shows a peak at 284 eV and a bulge around 291 eV, indicating the presence of amorphous carbon. The σ^* signal becomes more pronounced from the interior of the sphere to the edge of the sphere as far as the EELS detection is concerned, suggesting that the graphitization is more pronounced at the edge of the sphere location, which is consistent with prior observations of abundant NGs distributed at the edges of the nanospheres. The Si and O signals are weakened from the interior to the exterior of the sphere, while the C-K, S-L_{2,3}, and Mo-M_{4,5} signals are enhanced (as shown in the corresponding curves points 1 to 7). This gradient supports the hypothesis that the spheres have a SiO₂ core coated with a carbon film and wrapped with MoS₂. The EELS mapping provides a more intuitive display of this structure during friction. Combining the results of HADDF-STEM and BF-STEM, the microscopic features of the HFLC/MoS₂ nanosphere in the tribofilm were verified: MoS₂ was wrapped around the HFLC nanospheres under the action of friction, thereby forming the "HFLC/MoS₂ nanosphere" structure. The analysis of the friction interface morphology and the internal mechanism of the tribofilm, as well as the EELS examination of individual microspheres, similarly demonstrated the existence of the HFLC/MoS₂ nanosphere structure.

Consequently, it can be concluded that in the constructed MoS₂/HFLC nanosphere system, MoS₂ self-assembles and HFLC undergoes NG phase transition during friction. FL structures ruptured due to the shear strain and act as nucleation centers in the formation of nanocrystalline graphite [24]. Odd rings (five- and seven-membered rings) vanished to form nanocrystalline graphite stacks. This transforms the original shear interface into readily shearable interfaces including MoS₂/MoS₂, NG/NG and MoS₂/NG. Based on the multi-asperity contact mode, the combination of diverse shear interfaces gives rise to macroscale solid superlubricity.

To further clarify the formation mechanism of NG and reveal the superior shear performance of MoS₂/NG heterogeneous shear interfaces, molecular dynamics (MD) simulation was adopted to illustrate the transformation process from HFLC to NG (Fig. 8). To facilitate the observation of

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structural changes in HFLC, the shear extrusion process was monitored over a long period of time (Figs. 8a, b). At 0.8 seconds, the state diagram showed a mixed distribution of hexagonal and non-hexagonal rings, presenting an overall disordered state. Currently, the number of hexagonal carbon rings is small (six irregular hexagonal rings in the region at 0.800 seconds in Fig. 8c), with non-hexagonal rings being the predominant form. As the friction process progresses from 0.800 s to 1.600 s (Fig. 8c), non-hexagonal rings disintegrate and reorganize into hexagonal rings, increasing the number of hexagonal rings and enhancing the degree of order. Under shear extrusion, a short-range ordered NG structure is generated. Graphite (sp^2 carbon phase) is thermodynamically more stable than diamond (sp^3 carbon phase), with an energy barrier of 1.0 eV per atom [48]. Under shear stress, fullerene-like clusters disintegrate and serve as nucleation centers for nanocrystal formation. This friction-induced grain reconstruction process involves the disappearance of odd rings (five- and seven-membered) and the formation of stacked six-membered graphene sheets [49]. The resulting short-range ordered graphitic nanocrystals are distributed within an amorphous matrix, and the weak shear forces between them reduces friction and wear. Nucleation, film growth, and thickness saturation are essential for the formation of tribofilm, while nucleation centers are a prerequisite [50, 51]. Additionally, shear-induced "graphitization" is associated with an increase in sp^2 carbon bonds, forming weakly interacted hexagonal planes or compound nanocrystals through friction-induced reactions [52]. In summary, the transformation of fullerene-like structures into nanocrystalline graphite is driven by thermodynamic stability and facilitated by shear stress and friction and wear are reduced due to the formation of graphitic nanocrystals [24]. This indicates that HFLC can form NG structures under shear extrusion in which the number of hexagonal rings increases and stabilizes over time (Fig. 8d). This suggests that NG generated by friction can stably exist under shear extrusion conditions.

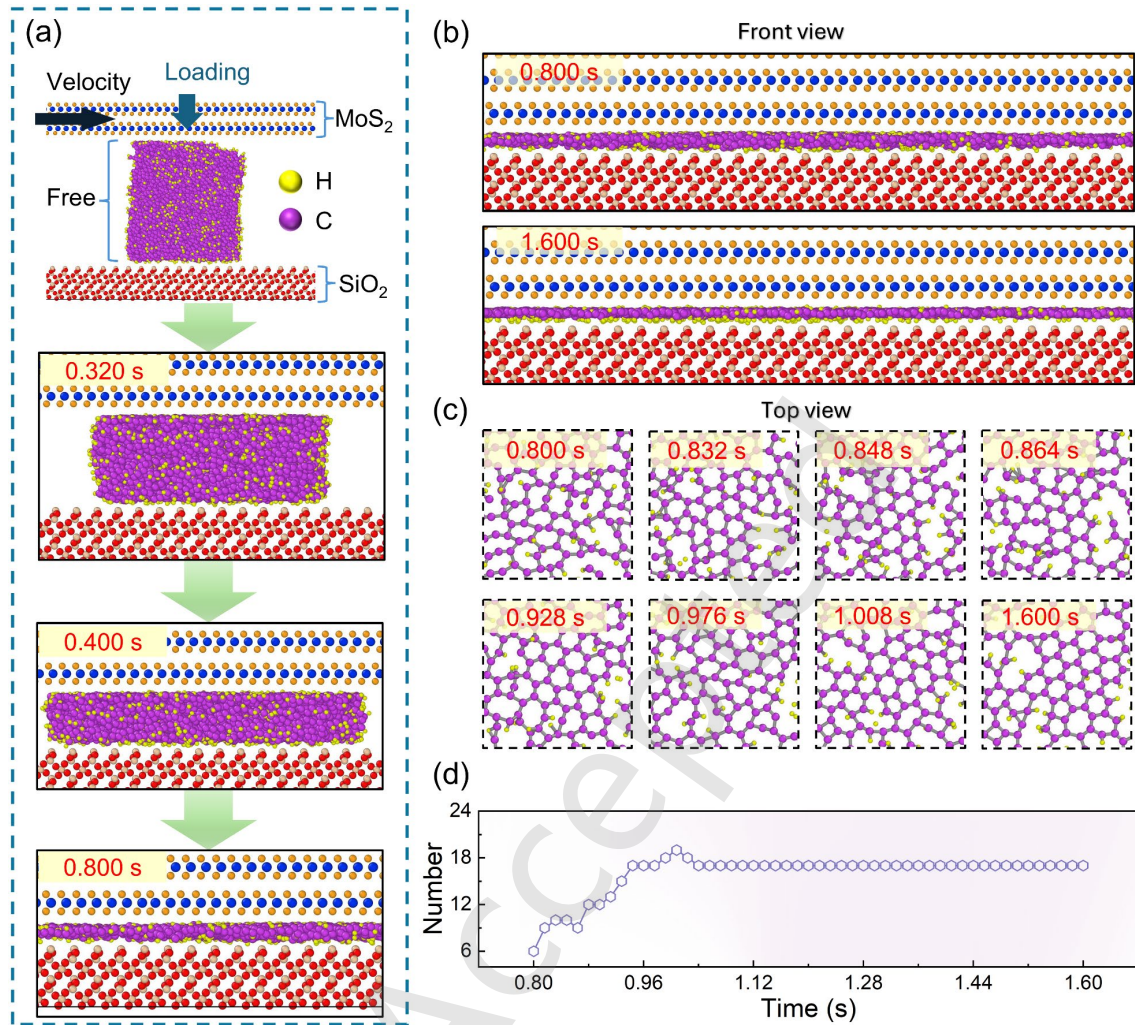


Fig. 8 MD simulations of the NG formation. a) Structure changes of HFLC in the established model from initial state to 0.800 s. b, c) Front view (b) and top view (c) of sixfold rings increasing process in HFLC structure from 0.800 s to 1.600 s. d) Statistics of the number of sixfold rings from (c).

Here, the sliding energy between MoS₂/NG layers (**Fig. 9a**) and MoS₂/MoS₂ layers (Fig. 9b) are compared to reflect the lubrication effect of the heterojunction. The relative energy along the sliding path of the MoS₂ layer in the X direction shows that the total energy of the MoS₂/NG interface remains almost constant. This indicates that MoS₂ slides easily on the carbon surface, with an energy barrier of 0.012 eV, which is significantly lower than the energy barrier of 0.532 eV on the MoS₂ surface (Fig. 9c). This phenomenon can be understood in terms of the incommensurate interface contact, where electronic interactions cancel each other out. There can be significant incommensurate contact between

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the MoS₂ and the carbon layers. These calculations demonstrate that the MoS₂/NG layer has better lubrication performance compared to the MoS₂/MoS₂ layer.

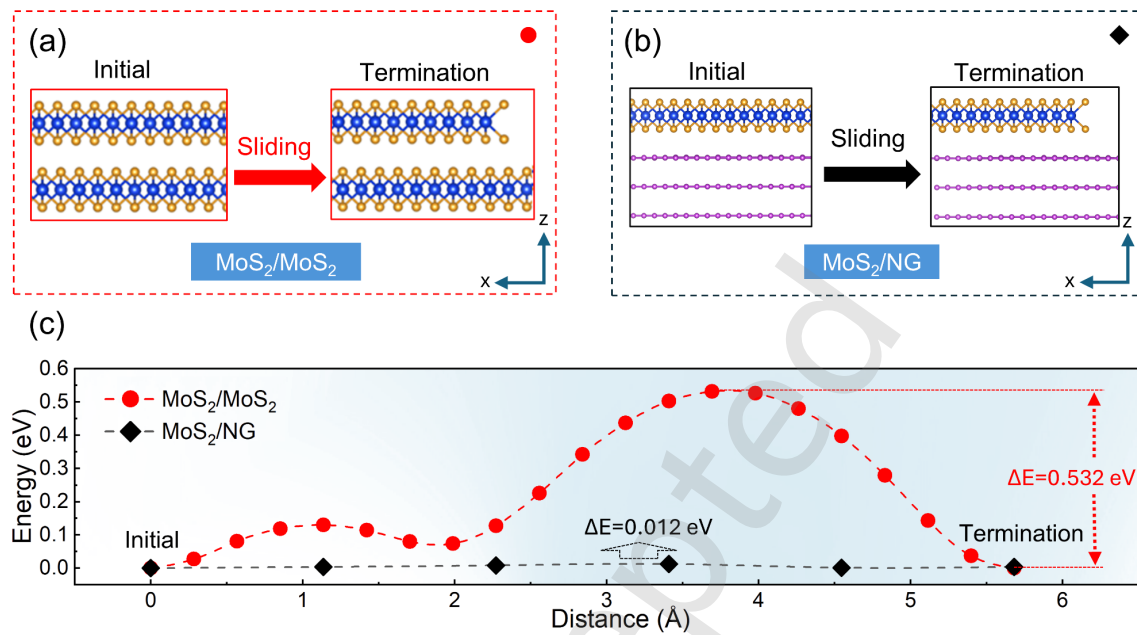


Fig. 9 Sliding energy barrier calculation. **a)** Initial and termination states of the MoS₂/MoS₂ calculation model. **b)** Initial and termination states of the MoS₂/NG heterojunction calculation model. **c)** Comparison of sliding energy fluctuation curves of MoS₂/MoS₂ model (red dot) and MoS₂/NG model (black rhombus) from initial state to termination state.

Through the comprehensive analysis of two surfaces and an interface, this study elucidates the fundamental factors contributing to the reduction in friction coefficient. The formation and accumulation of HFLC/MoS₂ nanospheres during the friction process is the main reason for the occurrence of superlubricity. Detection and analysis confirm that the nanospheres retain a spherical structure and form a new tribofilm (Fig. 10a). Nanospheres displace and reorient on the friction surface, facilitating the encapsulation of nanospheres by MoS₂ and continuous material replenishment, thereby maintaining the stability of the tribofilm (Fig. 10b). The new tribofilm reduces the actual contact area and further lowers the friction coefficient. During friction, MoS₂ interacts with the surface of the HFLC nanospheres to form NGs and a heterojunction. Simulation calculations verify the generation of NGs

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and demonstrate the heterojunction's contribution to the superlubricity phenomenon. Observations show that nanospheres divide the macroscopic interface into countless microscopic contacts, minimizing the impact of macroscopic defects on the friction coefficient. Experimental and test results confirm that numerous nanosphere structures can transform "single-asperity" macroscopic contact into "multi-asperity" microscopic contact, achieving macroscopic superlubricity through cumulative effects. Finally, a series of analyses demonstrates that the HFLC/MoS₂ nanosphere lubrication strategy of converting macroscopic contact to microscopic contact is feasible.

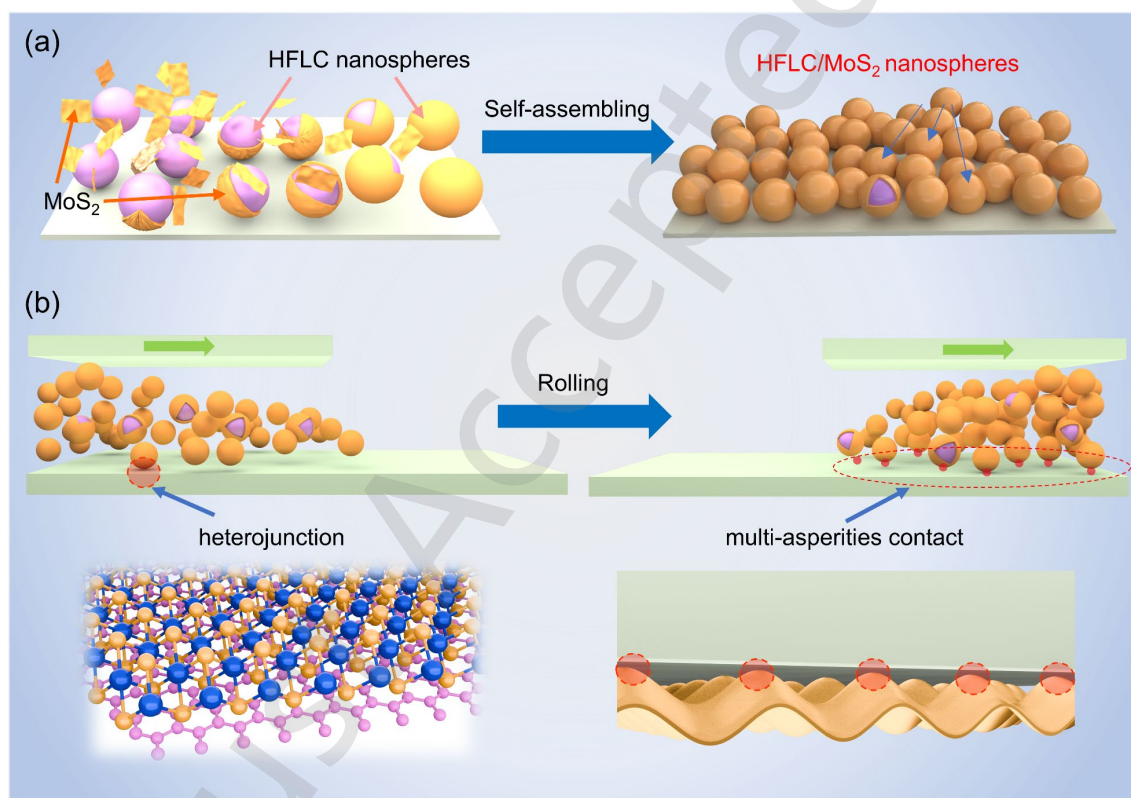


Fig. 10 Schematic illustration of the multi-asperity contact mechanism. a) Self-assembly of numerous microspheres to create a frictional interface, converting macroscopic surface contact into discrete microscopic point contacts. **b)** Induction of structural superlubricity at each point contact. This architecture effectively reduces contact area, mitigates chemical adhesion, and activates structural superlubricity at individual asperities via incommensurate sliding, ultimately achieving macroscale superlubricity.

4 Conclusions

This study validates a novel macroscale superlubricity strategy through the design and tribological analysis of the HFLC/MoS₂ nanosphere architecture. The proposed structure achieves macroscale superlubricity ($\mu = 0.009$) on steel substrates, maintaining stable performance ($\mu < 0.01$) under alternating vacuum and ambient air conditions. Experimental and simulation evidence reveals that the superlubricity arises from three synergistic mechanisms: (1) The spherical geometry provides a stable contact platform for the multi-point contact mode, thus maintaining the stability of the tribofilm; (2) Friction-induced formation of a tribofilm that fragments macroscopic single-asperity contact into microscopic multi-asperity interactions, thereby reducing the real contact area; (3) The formed MoS₂/NG heterojunctions ($\Delta E = 0.012$ eV) at the interface effectively suppresses interfacial adhesion. Finally, importantly, this superlubricity strategy — relying on the multi-asperity contact mode (also referred to as multi-point contact) to construct homogeneous and heterogeneous shear interfaces within the contact region — possesses broad theoretical applicability. The essence of this strategy lies in combining contact area reduction with easily slidable interlayer structures. Any self-assembled system that satisfies these two characteristics holds great potential for achieving superlubricity.

Funding

This work was supported by the National Natural Science Foundation of China (52475203, 52075309), the Innovation Support Plan of Shaanxi Province (2024ZC-KJXX-089), the Youth Innovation Team of Shaanxi Universities (2021–6), the Science and Technology Department, Shaanxi Province (2021GY–248), the Natural Science Foundation of Shaanxi Province (2023–JC–YB–428).

Author contributions

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Availability of data and materials

The datasets used and/or analysed during the current study are available from the corresponding author on reasonable request.

Competing interests

The authors have no competing interests to declare that are relevant to the content of this article.

Electronic Supplementary Material

Supplementary material is available in the online version of this article.

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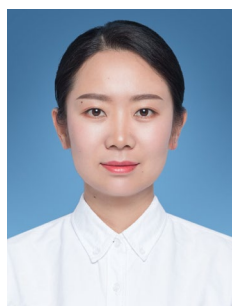
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