

Recent advances in friction and lubrication of graphene and other 2D materials: Mechanisms and applications

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Abstract: Two-dimensional materials having a layered structure comprise a monolayer or multilayers of atomic thickness and ultra-low shear strength. Their high specific surface area, in-plane strength, weak layer-layer interaction, and surface chemical stability result in remarkably low friction and wear-resisting properties. Thus, 2D materials have attracted considerable attention. In recent years, great advances have been made in the scientific research and industrial applications of anti-friction, anti-wear, and lubrication of 2D materials. In this article, the basic nanoscale friction mechanisms of 2D materials including interfacial friction and surface friction mechanisms are summarized. This paper also includes a review of reports on lubrication mechanisms based on the film-formation, self-healing, and ball bearing mechanisms and applications based on lubricant additives, nanoscale lubricating films, and space lubrication materials of 2D materials in detail. Finally, the challenges and potential applications of 2D materials in the field of lubrication were also presented.

Keywords: two-dimensional materials; graphene; anti-friction; anti-wear; lubrication

1 Introduction

Friction is a common phenomenon that is encountered in people's daily lives and in industrial production. It has been estimated that 1/2 to 1/3 of the world's energy is consumed in various forms of friction [1–3]. Friction causes wear and even failures in mechanical equipment. Therefore, the effective reduction of friction and the control of wear are important considerations in the performance of mechanical equipment and even in the improvement of the national economy.

The use of lubrication is an effective method of controlling friction and wear in mechanical equipment. Approximately 4,000 years ago, according to ancient Egyptian murals, water, animal products, and vegetable

oils were first used as lubricants to reduce friction, thereby increasing the service life of tools [4]. Liquid lubricants have been widely used owing to their effective anti-friction effect. However, the performance of liquid lubricants is seriously affected in severe working environments, which may result in the failure of the equipment. Thus, solid lubricants have been considered in the industry as they are more stable than liquid lubricants under the harsh conditions of heavy loads, high speeds, etc. As the micro- and nanoscale phenomena in tribology played an increasingly important role in modern technologies such as micro-electro-mechanical systems, traditional lubricants began to exhibit their limitations in the case of micro-scale friction. With the unparalleled superiority of

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traditional lubricants, the small size and surface effects of nanoparticles provide an excellent lubrication performance. Therefore, extensive studies on the friction properties and wear resistance of nanomaterials have been conducted by researchers in the field of tribology [5–8].

Two-dimensional (2D) materials are representative of new nanomaterials and are expected to introduce new opportunities for application in the traditional technology and engineering fields [9–19]. Atoms in the same atomic layer are combined through a covalent bond, which results in the formation of a monolayer structure having a high modulus and high strength. In addition, the low shear resistance between the adjacent atomic layers combined by the Van der Waals force allows the atomic layers to slide easily [20]. Because of the higher specific surface area of 2D materials, they are easily adsorbed onto the contact surface, which prevents the direct contact of the friction pair. Thus, the lubrication performance of 2D materials is superior to that of other nanomaterials. Graphene is a new type of 2D material with a hexagonal honeycomb 2D grid structure and comprising single-layer carbon atoms exhibiting sp^2 hybridization. The thickness of monolayer graphene is 0.335 nm [21]. The excellent tribological properties of 2D materials have also been widely explored. Cao et al. [22] focused on their mechanical properties including elastic modulus, strength, and fracture, and the frictional properties of graphene, MoS_2 and BN. In 2010, Lee et al. [23] studied the tribological properties of 2D materials such as graphene, MoS_2 , and BN. The experimental results they obtained showed that 2D materials have good anti-friction properties. Penkov et al. [24] reviewed the tribological properties of graphene and concluded that the key factors affecting tribological properties are the number of layers, stacking mode, and substrate material.

This review systematically describes the nanoscale friction mechanisms of 2D materials including the interfacial friction and surface friction mechanisms. ‘Superlubricity’ and three other major mechanisms including electron-phonon coupling effect, puckering mechanism, energy dissipation mechanism are described in detail. This review also introduces lubrication mechanisms based on the film formation mechanism,

self-healing mechanisms, and ball bearing mechanism and applications based on lubricant additives, nanoscale lubricating films, and space lubrication materials of 2D materials in detail. Finally, we present the challenges and prospects of 2D materials in this field.

2 Nanoscale friction mechanism of 2D materials

Despite great advances in science and technology and remarkable research progress on friction, the current understanding of friction and lubrication phenomena is macroscopic. Owing to the development of micro-mechanical devices, macroscopic tribology theory has become obsolete, and the characteristics of friction at the microscale have been extensively investigated [25, 26]. Tribology has also followed this developmental trend and has gradually evolved into nanotribology. Prof. Winer, a famous American tribology scholar, pointed out that microscopic and atomic scale tribology is a new field. Since the 1990s, several international scientific research teams have conducted numerous experiments using instruments such as scanning electron microscope (SEM), scanning tunneling microscope (STM), and atomic force microscope (AFM) in combination with molecular dynamics [27–32], first principle calculation [33–39], and finite element analysis [23, 40, 41]. They concluded that the friction mechanisms can be divided into inter-layer and surface-sliding friction mechanisms, which are explained in detail in the following sections (Fig. 1).

2.1 Sliding-friction mechanism of 2D material interlayers

2.1.1 Discovery of ‘superlubricity’

In 1928, Bragg [42] attributed the low-friction behavior of flake-like materials to the low shear resistance between their adjacent atomic layers. Tomlinson [43] used a well-known mechanical model to understand the mechanism of solid friction and revealed that solid sliding is caused by the dissipation of elastic energy owing to the relative sliding of two contacting solids. However, Tomlinson did not study the possible contact movement at a crystal surface at the atomic level. In 1990, Hirano et al. [44–47] studied two clean crystal

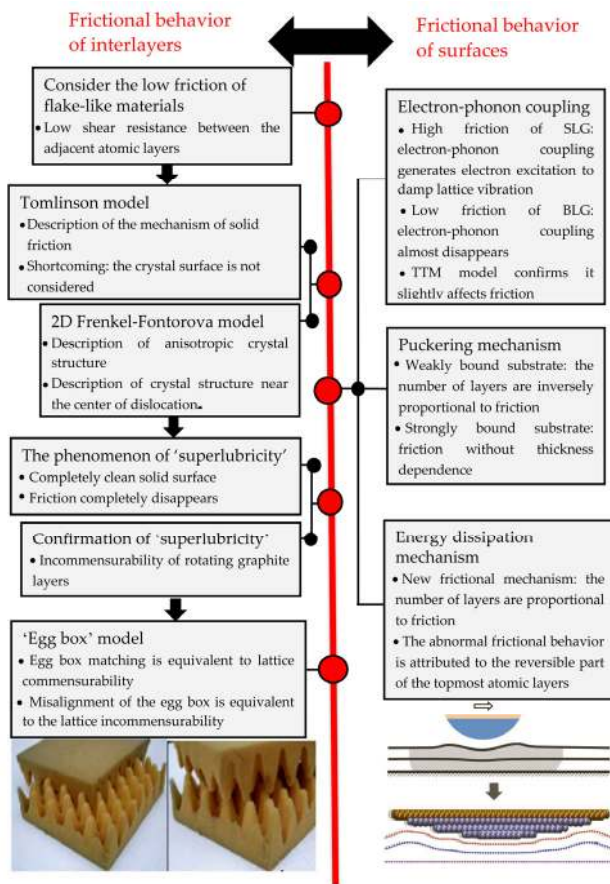


Fig. 1 Summary of frictional mechanisms of 2D materials.

surfaces and found special cases in which friction completely disappears at a completely clean solid surface. Subsequently, they confirmed the occurrence of the phenomenon called ‘superlubricity’ using ultrahigh vacuum STM. The presence of superlubricity depends on the commensurability of a contact surface. This conclusion was confirmed by Dienwiebel et al. [48, 49], who used a self-made friction microscope to examine the energy dissipation between sliding tungsten tips on a graphite surface by measuring the atomic friction as a function of the angle of rotation. They verified that the ultra-low friction of graphite was owing to the incommensurability of rotating graphite layers. Similarly, Martin et al. [50] studied the superlubricity of MoS₂ and attributed it to the incommensurability of its adjacent layers during intercrystallite slip.

To understand the concept of lattice commensurability, Zheng et al. [51] creatively developed the “egg box” model. It is equivalent to the perfect match of the lattice constants and the orientations of two crystal flakes when two egg boxes are completely

stuck, as shown in Fig. 1. Thus, the two flakes either exhibit commensurability or incommensurability.

Lattice commensurability dramatically affects the interlayer friction of graphene; ultra-low friction (superlubricity) occurs especially when surfaces come into contact incommensurately [52]. Moreover, these findings provided the basis for the further understanding of the friction mechanism between 2D material interlayers.

2.1.2 Interlaminar friction in homogeneous and heterogeneous sliding contact

To further explore the influence of the lattice commensurability of interlayers on the friction between layers, using STM, Feng et al. [53] found that graphene nanosheets slide easily on a graphene surface at temperatures as low as 5 K. This phenomenon includes translational and rotational motions in the initial and final states. In addition, they presented the sliding mechanism that occurs on graphene, which revealed the commensurate-incommensurate transition, as shown in Fig. 2. Furthermore, superlubricity may also occur in other 2D materials such as MoS₂ and BN because of their same layered structure. Li et al. [54] obtained a friction coefficient of 10⁻⁴ in the regime of superlubricity in the case of the sliding between incommensurate MoS₂ monolayers by combining the in situ SEM technique with a Si nanowire force sensor. The obtained results provided a new approach and guidance for studying the friction mechanisms of 2D materials, which was of great significance.

The registry index (RI) concept is used to quantify interlaminar copolymerization levels in layered materials using simple geometric considerations. An

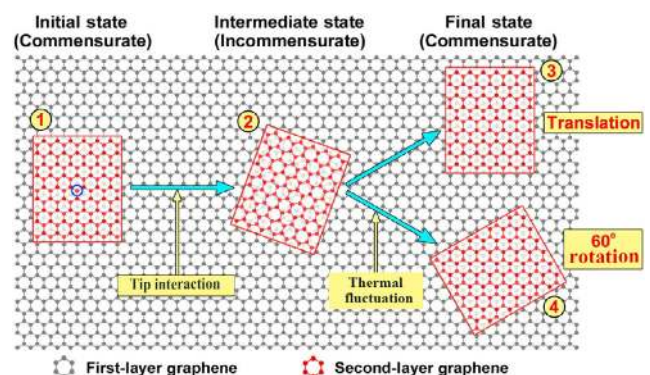


Fig. 2 Conversion process of commensurate-incommensurate state when graphene nanosheets slide on graphene surface. Reproduced with permission from Ref. [53], © American Chemical Society 2013.

accurate relationship between a commensurate interlayer and nonabrasive friction in layered materials has been presented by Hod, and its simple and intuitive model can be used to capture the behavior of hexagonal graphite sheets sliding on a graphite surface [55]. Furthermore, the ultra-high-speed superlubricity of micrometer-sized graphene flakes has been observed [56]. However, the superlubricity that occurs in finite-sized sheets is unstable. Experiments have shown that the low-friction sliding of an incommensurate graphite flake on graphite can be broken by rotation caused by torque, thus resulting in large and irreversible friction [57]. Superlubricity behavior easily develops into slip motion under high loads [58]. These findings have been supported by theoretical results. Wijn et al. [59] concluded that certain scanning lines, thermal fluctuations, and high load forces can destroy the stability of an ultra-lubricated track. The optimal conditions for superlubricity are a large layer, low temperature, and low load. In addition, Liu et al. [60] studied two methods of suppressing frictional scattering to maintain the state of superlubricity. One such method involves the use of graphene nanoribbons for eliminating the friction scattering by limiting the rotation of nanosheets, which are called frictional waveguides. Another such method includes the effective suppression of frictional scattering via the biaxial stretching of a graphite substrate.

In addition to effectively maintaining the superlubricity at a nanoscale, Berman et al. [61] found that superlubricity can be realized at an engineering scale. They concluded that nanoscrolls slide on a diamondlike carbon surface at which incommensurate contact occurs, thereby reducing the friction coefficient. Wu et al. [62] developed a lubricating system in which a self-assembled graphene film (SGF) slides against an SGF under macroscale contact. Ultra-low friction was discovered owing to the low resistance to shear between the adjacent layers of SGFs. In addition, they found that an annealing process would significantly enhance the tribological properties of SGFs. Another example is the macroscopic superlubricity exhibited between the inner and outer shells of centimeter-sized double-walled carbonnanotubes [63]. In general, the key to achieving stable superlubricity is to realize a sustained incommensurability sliding contact.

Liu et al. [64] achieved sustained superlubricity between layers in a high-load atmosphere. They prepared graphene-coated microspheres (GMS) using the metal-free catalyst chemical vapor deposition (CVD) method and obtained an ultra-low coefficient of friction of 0.0025 at a local rough-contact pressure of up to 1 GPa and an arbitrary relative surface rotation angle. This ultra-low friction is attributed to the incommensurate contact that can be realized between randomly oriented graphene nanocrystallites. The specific preparation of GMS is shown in Fig. 3. Vu et al. [65] investigated the friction forces in incommensurate micrometer-sized contacts between atomically smooth graphite surfaces. The ultra-low friction can be obtained in the case of a normal load (maximum pressure 1.67 MPa).

Superlubricity easily occurs when 2D materials slide on other 2D materials. This heterogeneous interlayer sliding-friction system has been extensively investigated and is expected to have a robust superlubricity behavior regardless of the orientation of the substrates [66–68]. The theoretical basis for this is that heterostructure materials naturally exhibit a lattice misfit.

RI theory—which is based on studies on the phenomenon of homogenous sliding structures—can accurately capture the frictional behavior of a hexagonal graphite flake on the surface of graphite and has been used to examine the sliding behavior of heterogeneous structures. Leven et al. [69] defined the RI of a graphene/h-BN interface and concluded that when sufficiently large graphene flakes slide on top of the h-BN layer, regardless of the relative orientation, a stable superlubricity state can be realized.

In addition to the lattice commensurability, the normal force and stacking structure between layers also influence superlubricity [70]. Moreover, graphite

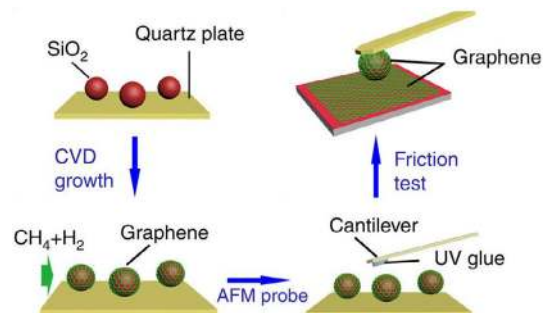


Fig. 3 Preparation of graphene-coated microspheres. Reproduced with permission from Ref. [64], © Springer Nature 2017.

contains different types of defects that affect the interlayer friction. The majority of theories and investigations mainly take into consideration perfect graphene sheets without defects [71, 72]. Guo et al. [73] examined the effect of interlayer distance variations and on-chip defects on inter-layer graphene friction by using a wide range of molecular field statics calculations. They found that the friction between the graphene layers increases as the interlayer distance decreases. In addition, the introduction of defects significantly affects the interlaminar friction in graphene with incommensurate stacking. The ultra-low friction of graphene layers stacked in an incommensurate manner is insensitive to the chip defects of a certain orientation of vacancies. It has been found that the friction between graphene layers is affected by various factors, such as temperature, load, size, stacking, defects, interlayer spacing, and the number of layers [74]. Xu et al. [75] investigated the effect of layer thickness on the intrinsic frictional properties of few layer graphenes and revealed the strong dependence of stick-slip friction force on the number of layers. As the number of layers decreases, the sliding friction gradually decreases. When there are only two or three atomic layers, the average friction is almost zero.

2.2 Sliding-friction mechanism of 2D material surfaces

In addition to its abundant interlayer sliding friction, the sliding friction of 2D materials on a graphene surface has been extensively investigated. This frictional mechanism is discussed in the following sections.

2.2.1 Electron-phonon coupling effect

The mechanism of friction at a sliding interface has yet to be elucidated because of the inherent complexity. Two basic mechanisms of energy dissipation owing to friction have been obtained: electron and phonon contributions [76, 77]. Filleter et al. [77] observed that the difference in friction between monolayer and bilayer graphene is attributed to the significant variation in electron-phonon coupling by using angle-resolved photoelectron spectroscopy. The possible explanation for the higher friction that occurs in monolayer graphene is that the lattice vibrations are efficiently

damped, and thus, the majority of the energy can be dissipated only through electron excitation. Dong et al. [78] applied molecular dynamic simulation and the two-temperature method (TTM) to confirm that electron-phonon coupling in graphene slightly affects friction as compared with the substrate roughness. Although electron-phonon coupling may be essential for atomic friction, simulations based on a TTM model show that friction is slightly related to electron-phonon coupling.

2.2.2 Puckering mechanism

The mechanisms of substrate morphology, electron-phonon coupling, and wrinkling are possible reasons for the thickness dependence of graphene friction, while the effect of electron-phonon coupling is weak. Thus, the state of the substrate and the puckering effect may seriously influence the dependence of the thickness of graphene friction. Lee et al. [23, 79, 80] found that the frictional force of atomically thin sheets obtained through mechanical exfoliation is closely related to the atomically thin-sheets–substrate binding state. When atomically thin sheets of graphene and MoS₂ are exfoliated onto a weakly adherent substrate (SiO₂/Si), the surface friction decreases as the number of layers increases. Suspended atomically thin sheet films exhibit the same properties. Friction is not affected by the number of layers when graphene is exfoliated onto the surface of fresh mica with a high adhesive strength. Thus, the state of the substrate has a significant influence on the friction force of 2D materials. As the probes slide on the surface of a weakly adherent substrate, the friction force is large because of the obvious wrinkle deformation of graphene. While the probes slide on the surface of fresh mica, the strong binding between graphene and fresh mica inhibits the wrinkle effect, and thus, the thickness dependence is not observed. Cho et al. [81] found that mechanically stripped graphene produces low friction on an atomically smooth substrate. This class of graphene inhibits surface wrinkle deformation as observed using atomic stick-slip imaging. This is because the conformal morphology of graphene on the substrate enhanced the intimate contact, increased the contact area, and suppressed the puckering effect, thereby resulting in

low friction. Thus, the influence of the morphology of the substrate is actually caused by the wrinkle effect.

This property is also validated in subsequent studies base on puckering mechanism. Furthermore, Deng et al. [82] found that the friction increases with increasing number of graphene layers at low loads and attributed this result to the local deformation of the surface layer and van der Waals forces between AFM tip and the surface layer. Ye et al. [83] observed that the influence of the number of layers on the friction becomes notable when the simulated graphene sheet exceeds 32 nm or longer. Frictional behavior can be directly related to the height of anti-slip approaching wrinkles, the bonding energy between graphene layers decreases as the size of graphene decreases, resulting in an increase in the distance between graphene layers which less able to resist wrinkle formation.

2.2.3 Energy dissipation mechanism

The layer dependence of the friction force has been extensively investigated. Smolyanisky et al. [84] conducted a Brownian dynamics (BD) simulation to quantify the friction behavior at room temperature between the scanning ends of carbon nanotubes and FLGs or the surface of self-supporting multi-layer graphene. They found that the friction decreases at 5 m/s as the number of layers increases, and this observation is the same as the conclusion presented by Lee et al. Subsequently, they investigated the contact area of different layers of graphene in the sliding process and found that the real contact area changes slightly. Therefore, the contact area between the tip and the sample is not the main reason for the change in the friction at the graphene surface with the number of layers. They proposed an energy dissipation mechanism based on the elastic energy of the specimen deformation. The energy dissipation mechanism of the deformation has been further explored. Deng et al. [85] discovered unusual frictional behaviors at the graphene surface. When a probe gradually departs from the surface of a graphene sheet, the friction force increases as the load under the tip contraction decreases, thus resulting in an effective negative coefficient of friction at a low load. The size of this coefficient depends on the ratio of the adhesion of the tip-sample to the peel energy of graphite, and this

unusual phenomenon is attributed to the delamination of the reversible part of the topmost atomic layer. A probe lift-off or applied load reduction results in a considerable surface deformation, thus increasing the frictional force (Fig. 4).

Although the energy dissipation mechanism remains unverified, the influence of lateral slip is important. Sun et al. [86] used molecular dynamics simulations to study the lateral sliding behavior of graphene. Their results revealed that the energy corrugation associated with sliding is not only a result of the interfacial interaction but also the interaction between the atomic deformation layers of graphite. In addition, the unusual phenomenon that occurs during the tip retraction was correlated with the local atomic

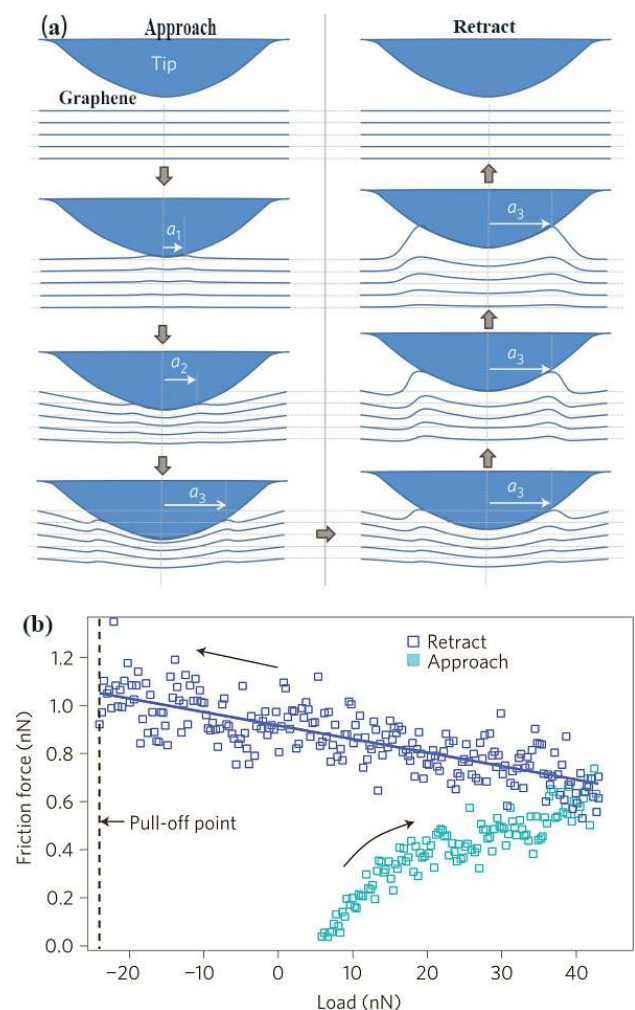


Fig. 4 (a) Schematic of retraction when tip approaches and leaves graphene surface; (b) unusual frictional behavior at graphene surface. Reproduced with permission from Ref. [85], © Springer Nature 2012.

delamination of the top graphene layers (Fig. 5). This is consistent with the anomalous experimental phenomenon observed by Deng et al. at the surface of graphene [85].

Reguzzoni et al. [87] used molecular dynamics simulations to investigate the tribological properties of multilayer graphene under energy dissipation due to shear deformation. They proposed a new friction mechanism related to shear motion, in which the friction force decreases as the number of layers decreases.

In summary, whether in theory or in an experiment, the nanoscale friction mechanisms of 2D materials have been extensively studied and have become one of the most popular subjects of study. A plane formed by a strong chemical bond between 2D materials has high in-plane strength, high surface chemical stability, and weak van der Waals force between its layers. Therefore, many strange friction mechanisms occur between the layers and surfaces. At present, although the research methods used vary widely, the results obtained are generally consistent; that is, 2D materials are expected to be promising nanoscale lubricating materials owing to their low friction coefficient and high load-bearing capacity. When 2D materials are used as additives in various types of macro-lubrication systems, protective films are often formed on the friction surfaces (improving surface roughness, repairing wear, bearing loads, preventing contact, providing low shear strength between layers, and causing interlaminar sliding) with nanoscale superlubricity properties. Therefore, they have important applications in the fields of lubricants, lubricating coatings, lubricating films, and space lubrication materials.

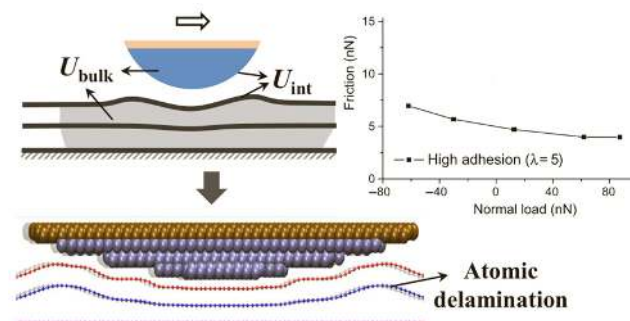


Fig. 5 Local atomic delamination of graphene and abnormal variation of friction with a normal load. Reproduced with permission from Ref. [86], © Springer Nature 2015.

3 Nanoscale lubrication mechanism of 2D materials

The lubrication mechanism should be studied in order to fully reveal the tribological properties of the nanoparticles. 2D materials provide interlaminar sliding and a low shear force, thus giving them superlubricity properties. Moreover, owing to their nanostructures, they can easily enter the frictional contact surface to form a lubricating film. Furthermore, they can also decrease surface roughness and repair wear. However, the elucidation of this mechanism remains the subject of many debates in research on nanoparticle lubrication systems. Researchers have described the mechanism of lubricant enhancement using nanoparticle suspensions via surface analysis techniques, such as ball bearing mechanism [88–92], film formation mechanisms [93–98] and self-healing mechanisms [99–102]. These mechanisms can be categorized into two broad categories. One category comprises the direct nanoparticle effects, including the ball effect and film formation. The other category comprises the assisting effect of surface enhancement via the repair and polishing effect. These mechanisms are discussed in the following sections.

3.1 Film formation mechanism

Nanoparticles with large surface areas show chemical activities and become quickly adsorbed onto a friction surface to form a physical adsorption film. Some nanoparticles are affected by external factors in terms of friction surface migration and deposited on a friction surface to form a deposited film. Nanoparticles can also react chemically at the friction surface to create a chemical reaction film, thereby enhancing the wear resistance of the friction-pair surface [103]. Hu et al. [104] studied the friction and wear properties of nano-MoS₂/TiO₂ nanoclusters and found that the MoS₂ frictional surface contains chemical films such as MoO₃, Fe₂O₃ (or Fe₃O₄), Fe₂(SO₄)₃ (or FeSO₄), FeS, and carbon-containing compounds. Therefore, they speculated that nano-sized TiO₂ and Mo are transferred onto the surface of the nano-MoS₂/TiO₂ nanocluster via a friction chemical reaction and react with other

substances to produce a lubricating film that reduces the surface friction and wear. Su et al. [105] observed the lubricating wear scar surface of 0.25% graphite nanoparticles added to plant-based oil. Graphite nanoparticles can be stably adsorbed on friction surface, which forms a physical adsorption film on a friction surface. Therefore, they speculated that graphite nanoparticles are mainly used to improve the anti-friction and anti-wear properties of plant-based oils because graphite nanoparticles can create physically deposited films on a surface.

Based on this theory, Xiao et al. [106] stated that the film formation process can be divided into two stages. The initial high specific surface allows 2D materials to be easily adsorbed onto the surface of a substrate to form a physical membrane. The physical membrane can separate the two contact surfaces to prevent direct contact between the two sliding surfaces. In the second stage, the physical film ruptures with an increase in the frictional strength, thereby promoting the chemical reaction between a lubricant and a local contact surface. This chemical reaction forms a new tribological film and gradually replaces the physical film, and this film exists on a local contact surface. As a result, the tribological properties are improved.

3.2 Self-healing mechanism

With inherent limitations in manufacturing technology, the contact surface remains significantly rough. Nanomaterials can fill a concave area on a friction surface to smoothen it. Su et al. [105] developed a model for simulating the self-healing mechanisms of nanomaterials (Fig. 6). The friction surface is tightly bonded under a high pressure, thus resulting in friction and wear. When an amount of nanomaterials is added to the lubricating oil to penetrate the concave

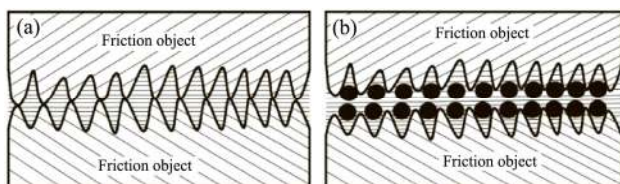


Fig. 6 (a) Schematic of a tribological model of lubricating base oil; (b) schematic of a tribological model of lubricating base oil containing an appropriate amount of nanomaterials. Reproduced with permission from Ref. [105], © Hindawi Publishing 2015.

friction surface, the damaged surface is repaired. The instantaneous high temperature during sliding can even melt the nanoparticles and repair defects on the sliding surface. Similarly, Gulzar et al. [107] investigated the self-healing effect of nanomaterials. Nanoparticles deposit on interacting surfaces and compensate for the mass loss, thereby reducing wear and tear.

3.3 Ball bearing mechanism

Nanoparticles disperse through friction repair to form “class bearings” and transform sliding friction into rolling friction, thereby reducing the friction coefficient and exhibiting an excellent anti-friction performance. Gulzar et al. [107] established a mechanism model to study the class bearing effect of nanomaterials and found that nanoparticles transform sliding friction into a combination of sliding friction and rolling friction. This lubrication mechanism is attributed to a friction pair system with a stable low-load condition between the shear surfaces to maintain the shape and stiffness of the nanoparticles (Fig. 7).

Xiao et al. [106] summarized the lubrication mechanism of 2D materials into four processes (Fig. 8) and proposed other mechanisms. For example, (1) 2D materials easily enter the friction pair because of their small size, and the relative movement of the contact surfaces results in a shear force acting on these materials. Consequently, multi-layered 2D materials are easily cut, and they readily come in contact with a friction surface to form a sliding system, thus resulting in effective lubrication. For instance, a MoS_2 sheet with lubricating grease comes in contact with the friction surface between two friction pairs. The sheet structure is easily absorbed onto the friction surface because of the lamellar crystal structure of MoS_2

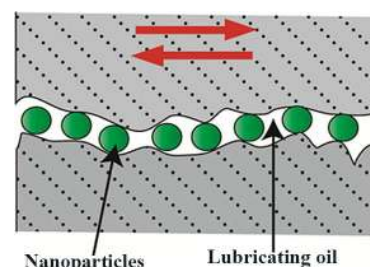


Fig. 7 Schematic of ball bearing mechanism of nanoparticles. Reproduced with permission from Ref. [107], © Springer Nature 2016.

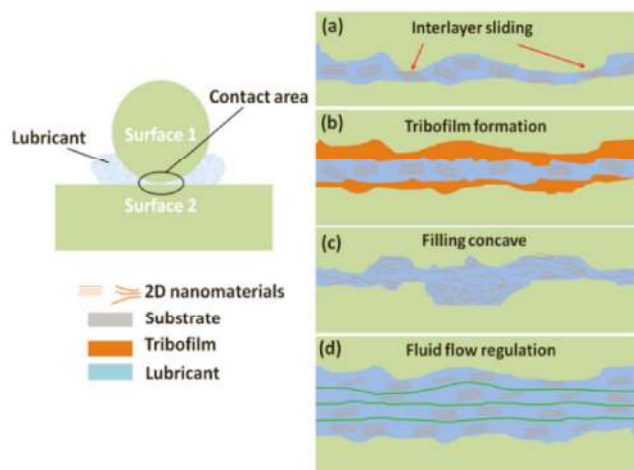


Fig. 8 Lubrication mechanisms of 2D materials. (a) entering the contact area of sliding surfaces, (b) tribofilm formation, (c) filling the pits and gaps of the contact area, (d) affecting the fluid drag and viscosity. Reproduced with permission from Ref. [106], © Elsevier 2017.

and the combined van der Waals forces between the layers. Under the effect of the frictional shearing force and normal load, cleavage easily occurs between the lamellar MoS_2 layers. Cleavage is also observed along the cleavage plane slip, which plays an anti-friction role. (2) 2D materials are impervious to liquids and gases because of their ultra-high chemical stability. The adsorption of these materials on substrates helps to prevent chemical attacks on the lubricants or other active elements in a given system, thereby slowing the corrosion and oxidation of materials and further reducing the wear on the sliding surfaces.

4 Application of 2D materials in lubrication

2D materials have excellent friction behavior mechanisms, and they have a high specific surface area (cover a greater surface area), high in-plane strength (bond by covalent bonding), good surface chemical stability, and low interlaminar shear strength (allow adjacent layers to slide easily against each other). Therefore, their surface lubrication effect is apparent, which is very suitable for applications in the lubrication field, such as lubricant additives, space lubrication materials, and nanoscale lubricating films.

4.1 Lubricant additives

Given their unique molecular structure and lubrication

properties, 2D materials are widely used in the field of tribology. They are also excellent candidates for the lubrication of friction surfaces because of their high strength and the low interlaminar shear strength between their atoms. The anti-friction and anti-wear performance of 2D materials is superior to that of conventional lubricant additives. In addition, 2D materials can also be used to reduce the emission of harmful substances and make an important contribution to building a green environment [108–111].

2D materials are widely used as oil-based lubricants. Zhao et al. [112] used a UTM-2 friction and wear testing machine for a graphene abrasion resistance test, and the obtained results demonstrated that graphene can significantly improve the friction and wear properties and even reduce the maximum friction coefficient by 78%. Furthermore, the maximum wear rate can be decreased by 95%. Senatore et al. [113] studied the tribological behavior of graphene oxide nanosheets in mineral oil. The experimental results indicated that the average friction coefficient decreases by approximately 20%, and the anti-wear rate decreases by approximately 30% at 25 °C – 80 °C and an average contact pressure of 1.17 GPa. The friction reduction mechanisms of graphene and graphene oxide are similar. They easily form a protective film to prevent the direct contact between the contact surfaces. In addition, the nanoscale thickness and extremely thin laminated structure offer a lower shear strength, thereby easily causing interlayer sliding and resulting in a lower friction.

Although graphene can effectively play anti-friction and anti-wear roles, graphene is prone to agglomerating in the base lubricating oil, which seriously affects the lubricating effect. In order to avoid the agglomeration of graphene, (1) an appropriate amount of dispersant is used to improve the uniform dispersion in the lubricating oil, and (2) an appropriate chemical modification (such as fluorination or hydrogenation) of graphene is used to enhance the uniform dispersion of graphene in the lubricating oil. Zhang et al. [114, 115] utilized the modification effect of an oleic acid surfactant to disperse graphene evenly and uniformly in poly- α -olefin (PAO9) lubricating oil and used a four-ball abrasion testing machine to test the anti-friction and anti-wear performance of modified graphene (Fig. 9). The obtained experimental results

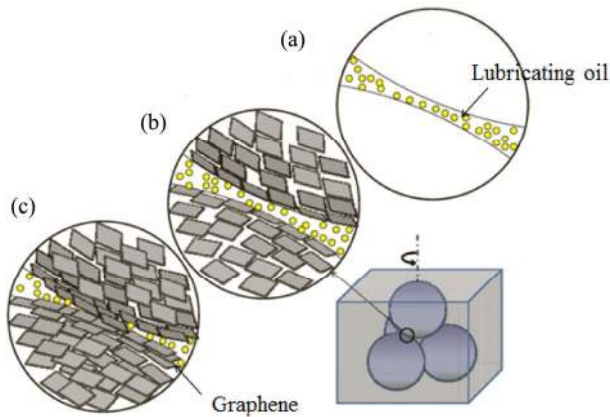


Fig. 9 Schematic of tribological experiment with lubricating oil adding a certain amount of graphene. (a) friction diagram of pure lubricating oil, (b) represents the optimal concentration of graphene in oil, (c) represents excessive concentration of graphene in oil. Reproduced with permission from Ref. [115], © IOP Publishing 2011.

show that the friction coefficient decreases by 17% when the mass fraction of the oleic-acid-modified graphene is 0.02%, whereas the wear spot diameter is reduced by 14% when the mass fraction is 0.06%. In addition, they concluded that an appropriate quality of graphene is required in the lubrication in order to achieve a friction reducing effect.

Lin et al. [116] conducted studies on the role of such modifiers and found that graphene is effectively modified by stearic acid and oleic acid, which is uniformly dispersed in the lubricating oil as an additive. The wear resistance and bearing capacity of the lubricating oil are significantly improved. This is because long paraffin chains on the surface of graphene produce a steric hindrance when modified graphene is dispersed in the base lubricating oil to prevent graphene sheets from being precipitated and agglomerated. Considering that the modification effect can obviously improve the dispersibility of graphene, Zheng et al. [117] applied ball milling to prepare graphene fluoride to improve the bearing capacity and wear resistance of lubricants, and the thus obtained base oil had good dispersion stability. In addition, they noted that fluorinated graphene sheets can easily enter the contact surface and form a protective layer, which reduced the wear and improved the load carrying capacity. However, when the concentration exceeds a critical value, the excess fluorinated graphene sheets might agglomerate with the metal wear debris, thereby

reducing the wear resistance. The conclusion of this study has been confirmed. Dou et al. [118] used crumpled graphene balls as additives to polyalphaolefin base oils. Owing to the unique self-dispersibility of crumpled graphene balls, they reduce the friction coefficient and wear coefficient by approximately 20% and 85%, respectively. In summary, to realize the wide applications of 2D materials in the field of lubrication, the problem of agglomeration is required to be further studied.

2D materials also act as water-based lubricating additives to maintain excellent thermal conductivity and the bearing capacity of water-lubricated films. Song et al. [119] investigated the tribological properties of graphene oxide nanosheets as water-based lubricating additives by using a UMT-2 ball friction tester. The result showed that graphene oxide nanosheets slide between the friction surfaces and form a thin physical friction film on the friction surface, which can bear friction and prevent the direct contact of the steel ball surface. Cho et al. [120] studied the lubricating effect of the water dispersions of *h*-BN nanoplates. They synthesized *h*-BN aqueous dispersions at concentrations of 1%, 0.05%, and 0.01% and evaluated the friction and wear of the aqueous dispersions using SiC balls sliding on a disk. They demonstrated that even a small amount of *h*-BN nanosheets can enhance the wear resistance and reduce the friction. A repeated peeling and deposition of *h*-BN occurs on the sliding surface, thereby forming a protective film that can reduce friction and wear. He et al. [121] studied the lubricating effect of α -zirconium phosphate in both oil and aqueous media using a characterization technique, and revealed that they are effective lubricant additives in oil and aqueous media. Subsequent to the addition of them in mineral oil and water, the friction is reduced by 65% and 91%, respectively. Their unique 2D structure promotes the arrangement of zirconium phosphate nanosheets in lubricants, thereby resulting in effective lubrication.

4.2 Space lubrication materials

Sputtered MoS₂ solid lubricants are widely used in the aviation field, e.g., high-speed long-life bearings for gyroscopes and accelerometers and gears and bearings for spacecraft harmonic drive devices [122].

Magnetron sputtering technology provides numerous advantages, including a high film-formation rate, low deposition temperature, uniform film thickness, and dense film structure. This technology has been applied for preparing various wear-resistant, corrosion-resistant, and anti-oxidation coatings and solid lubricant coatings [123, 124]. Numerous practical applications of MoS₂ sputtering films have been developed, and using the Aerospace 510, it has been shown that the service life of these sputtering films can exceed times, and individual test results have reached times [125].

Extreme conditions such as vacuum, low temperature, and strong radiation in the space environment demand special performance requirements of solid lubricants [126]. To satisfy the high reliability requirement of moving parts in space equipment and extend their life expectancy, Cheng [127] used nanoscale MoS₂ as an additive for space grease in an atmospheric environment and a simulated space environment, experimentally studied the anti-friction and anti-wear effects of the grease, and concluded that the friction and wear resistance of nano-MoS₂ are superior to those of MoS₂. Space grease with MoS₂ nanosheets has the best extrusion performance and anti-friction and anti-wear properties and good lubrication performance under high-load conditions. However, an excessive amount of nano-MoS₂ affects the flow of space grease, thus resulting in significantly decreased lubrication. Luo [128] experimentally concluded that the addition of nano-MoS₂ particle/polyester polymer aviation lubricants significantly improves the anti-wear performance, and the optimal concentration is 3 wt% to 4 wt%. Song et al. [129] found that graphene can solve the problem of vacuum lubrication and proposed a unique mechanism. They introduced graphene into a carbon film and grew 2D nanostructured carbon-based film materials on a carbon film using a field-induced growth method. An ultra-low coefficient of friction of 0.02 was thus obtained. The graphite surface is disorderly tiled, which impedes relative motion, thus resulting in a large friction coefficient. A graphene sheet has π electrons and certain surface interaction forces at the 2D structure surface and can form an oriented layered structure. Only a weak van der Waals force acts between its layers, which makes it prone to slipping, thus allowing it to exhibit ultra-low

frictional characteristics with nanoscale superlubricity properties.

4.3 Nanoscale lubricating film

Graphene-layered structures have an ultra-thin thickness, ultra-low shear strength, high chemical stability, and high specific surface area; thus, they are suitable for use in micro/ nanofilm equipment. Lee et al. [130] deposited graphene onto a copper surface through CVD to form a thin film, and the friction coefficient of a copper film deposited with graphene is lower than that of bare copper foil. Watanabe et al. [131] prepared a WS₂/MoS₂ nanocomposite film, i.e., a superlattice structure multilayer film using radio frequency sputtering. The multilayer films exhibit a wear resistance superior to that of individual monolayers. This multilayer film shows potential for the improvement of wear resistance relative to current MoS₂-based low-friction films and may be suitable for use under severe frictional conditions because of the increase in film hardness caused by the multilayer structure. Kim et al. [132] discovered the excellent frictional properties of graphene films grown on Cu and Ni metal catalysts and transferred the two graphene films to SiO₂/Si substrates through the CVD method. It was found that graphene films effectively reduce the adhesion and friction force. Graphene grown on Ni has a friction coefficient of 0.03, and the frictional difference is the appearance of a tortoise-like amorphous carbon layer on the Ni-grown graphene. In general, CVD-grown graphene films exhibit a strong potential for reducing adhesion and friction and protecting the substrate surface. The liquid phase exfoliation method can be used to produce high-yield graphene films, while the film performance obtained using this method is degraded owing to the functional groups and structural defects. Sun et al. [133] presented an annealing method in which nickel sputtering is used for the structural repair of graphene sheets by stitching the spacers, thus forming a continuous film, which improves the crystal quality. Similarly, Li et al. [134] performed thermal annealing on copper foils, coalesced the electrochemically exfoliated graphene flakes, and finally recrystallized it into a continuous film. The new growth mechanism of this recrystallized and coalesced graphene can be used to prepare a

high-quality graphene film having a large area, which has wide applications.

5 Summary and outlook

2D materials exhibit anti-friction and anti-wear properties because of their unique layered structure, and thereby providing great prospects for the development of nanoscale lubricants. In this paper, we reviewed the friction mechanism of 2D materials and found that this mechanism can be divided into the interfacial friction and surface friction mechanisms. The interfacial friction mechanism is mainly influenced by temperature, load, size, stacking, defects, layer spacing, and the number of layers, while the surface friction mechanism is mainly categorized into the electron-phonon coupling effect, out-of-plane puckering mechanism, and deformation energy dissipation mechanism. The lubrication mechanism includes the film formation mechanism, ball bearing mechanism, self-healing mechanism, and other lubrication mechanisms. In practical applications, 2D materials are commonly used as lubricant additives, nano-lubricating films, and vacuum space lubricating materials because of their unique anti-friction and anti-wear properties. Finally, to facilitate the use of 2D materials, we opine that future studies should be performed in the following areas:

1) The study of atomic level friction is a new direction in the field of tribology. Research on superlubricity may help to promote the development of the industry and energy, and thus, further research is required for achieving superlubricity under macro- and micro-scale conditions and maintaining the durability of superlubricity [135, 136].

2) Although 2D materials have excellent frictional properties, they are easily affected by the surface conditions, thus resulting in a degraded performance. Therefore, the optimization of various processes is inevitable during the process of preparation and the experiment.

3) Functionally modified 2D materials have superior friction properties owing to the inhibition of the agglomeration of graphene. Therefore, the preparation of functional 2D materials and the mechanism of the suppression of agglomeration should be further studied.

4) 2D materials quickly adsorb onto the surfaces of the friction pair because of their high specific surface area, and further chemical reactions occur. Therefore, further study is required to understand those physical and chemical reactions in order to make the lubrication mechanism of 2D materials more reliable.

5) 2D non-lamellar materials have great advantages in energy conversion and storage in the electronics industry because of their complex electronic structure [137, 138].

6) The preparation of new materials in the 2D materials family and the physical and chemical properties of the materials require further exploration.

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References

- [1] Wen S Z, Huang P. *Principles of Tribology*. 4th ed. Beijing (China): Tsinghua University Press, 2012.
- [2] Holmberg K, Andersson P, Erdemir A. Global energy consumption due to friction in passenger cars. *Tribol Int* 47: 221–234 (2012)

- [3] Holmberg K, Andersson P, Nylund N O, Makela K, Erdemir A. Global energy consumption due to friction in trucks and buses. *Tribol Int* **78**: 94–114 (2014)
- [4] Nosonovsky M. Oil as a lubricant in the ancient middle east. *Tribol Online* **2**(2): 44–49 (2007)
- [5] Berman D, Erdemir A, Sumant A V. Graphene: A new emerging lubricant. *Mater Today* **17**(1): 31–42 (2014)
- [6] Le Cao Ky D, Tran Khac B C, Le C T, Kim Y S, Chung K H. Friction characteristics of mechanically exfoliated and CVD-grown single-layer MoS₂. *Friction* **6**(4): 395–406 (2018)
- [7] Spear J C, Ewers B W, Batteas J D. 2D-nanomaterials for controlling friction and wear at interfaces. *Nano Today* **10**(3): 301–314 (2015)
- [8] Li Q Y, Zhang S, Qi Y Z, Yao Q Z, Huang Y H. Friction of two-dimensional materials at the nanoscale: Behavior and mechanisms. *Chin J Solid Mech* **38**(3): 189–214 (2017)
- [9] Chen Z F, Wang Z, Li X M, Lin Y X, Luo N Q, Long M Z, Zhao N, Xu J B. Flexible piezoelectric-induced pressure sensors for static measurements based on nanowires/graphene heterostructures. *ACS Nano* **11**(5): 4507–4513 (2017)
- [10] Li J J, Ge X Y, Luo J B. Random occurrence of macroscale superlubricity of graphite enabled by tribo-transfer of multilayer graphene nanoflakes. *Carbon* **138**: 154–160 (2018)
- [11] Li X M, Lv Z, Zhu H W. Carbon/silicon heterojunction solar cells: State of the art and prospects. *Adv Mater* **27**(42): 6549–6574 (2015)
- [12] Li X M, Zhu H W, Wang K L, Cao A Y, Wei J Q, Li C Y, Jia Y, Li Z, Li X, Wu D H. Graphene-on-silicon schottky junction solar cells. *Adv Mater* **22**(25): 2743–2748 (2010)
- [13] Li X M, Zhu M, Du M D, Lv Z, Zhang L, Li Y C, Yang Y, Yang T T, Li X, Wang K L, et al. High detectivity graphene-silicon heterojunction photodetector. *Small* **12**(5): 595–601 (2016)
- [14] Ouyang W G, Ma M, Zheng Q S, Urbakh M. Frictional properties of nanojunctions including atomically thin sheets. *Nano Lett* **16**(3): 1878–1883 (2016)
- [15] Wang L F, Ma T B, Hu Y Z, Wang H, Shao T M. Ab initio study of the friction mechanism of fluorographene and graphene. *J Phys Chem C* **117**(24): 12520–12525 (2013)
- [16] Xu Q, Li X, Zhang J, Hu Y Z, Wang H, Ma T B. Suppressing nanoscale wear by graphene/graphene interfacial contact architecture: A molecular dynamics study. *ACS Appl Mater Interfaces* **9**(46): 40959–40968 (2017)
- [17] Yang M M, Zhang Z Z, Zhu X T, Men X H, Ren G N. *In situ* reduction and functionalization of graphene oxide to improve the tribological behavior of a phenol formaldehyde composite coating. *Friction* **3**(1): 72–81 (2015)
- [18] Zhao G K, Li X M, Huang M R, Zhen Z, Zhong Y J, Chen Q, Zhao X L, He Y J, Hu R R, Yang T T, et al. The physics and chemistry of graphene-on-surfaces. *Chem Soc Rev* **46**(15): 4417–4449 (2017)
- [19] Zheng X H, Gao L, Yao Q Z, Li Q Y, Zhang M, Xie X M, Qiao S, Wang G, Ma T B, Di Z F, et al. Robust ultra-low-friction state of graphene via moiré superlattice confinement. *Nat Commun* **7**: 13204 (2016)
- [20] Onodera T, Morita Y, Suzuki A, Koyama M, Tsuboi H, Hatakeyama N, Endou A, Takaba H, Kubo M, Dassenoy F, et al. A computational chemistry study on friction of h-MoS₂. Part I. Mechanism of single sheet lubrication. *J Phys Chem B* **113**(52): 16526–16536 (2009)
- [21] Wang W P, Qu W S, Zhao J G. Preparation and lubrication performance of soluble graphene. *Carbon Tech* **34**(6): 17–20 (2015)
- [22] Cao C H, Sun Y, Filleter T. Characterizing mechanical behavior of atomically thin films: A review. *J Mater Res* **29**(3): 338–347 (2014)
- [23] Lee C G, Li Q Y, Kalb W, Liu X Z, Berger H, Carpick R W, Hone J. Frictional characteristics of atomically thin sheets. *Science* **328**(5974): 76–80 (2010)
- [24] Penkov O, Kim H J, Kim H J, Kim D E. Tribology of graphene: A review. *Int J Precis Eng Manuf* **15**(3): 577–585 (2014)
- [25] Liu Z, Yang J R, Grey F, Liu J Z, Liu Y L, Wang Y B, Yang Y L, Cheng Y, Zheng Q S. Observation of microscale superlubricity in graphite. *Phys Rev Lett* **108**(20): 205503 (2012)
- [26] Marchetto D, Feser T, Dienwiebel M. Microscale study of frictional properties of graphene in ultra high vacuum. *Friction* **3**(2): 161–169 (2015)
- [27] Ansari R, Ajori S, Motevalli B. Mechanical properties of defective single-layered graphene sheets via molecular dynamics simulation. *Superlattices Microstruct* **51**(2): 274–289 (2012)
- [28] Smolyanitsky A, Killgore J P. Anomalous friction in suspended graphene. *Phys Rev B* **86**(12): 125432 (2012)
- [29] Yoon H M, Jung Y, Jun S C, Kondaraju S, Lee J S. Molecular dynamics simulations of nanoscale and sub-nanoscale friction behavior between graphene and a silicon tip: Analysis of tip apex motion. *Nanoscale* **7**(14): 6295–6303 (2015)
- [30] Hu J N, Ruan X L, Chen Y P. Thermal conductivity and thermal rectification in graphene nanoribbons: A molecular dynamics study. *Nano Lett* **9**(7): 2730–2735 (2009)

- [31] Kang J W, Lee K W. Oscillatory behavior of graphene nanoflake on graphene nanoribbon. *J Nanosci Nanotechnol* **15**(2): 1199–1202 (2015)
- [32] Sasaki N, Okamoto H, Itamura N, Miura K. Atomic-scale friction of monolayer graphenes with armchair- and zigzag-type edges during peeling process. *e-J Surf Sci Nanotechnol* **8**: 105–111 (2010)
- [33] Kwon S K, Ko J H, Jeon K J, Kim Y H, Park J Y. Enhanced nanoscale friction on fluorinated graphene. *Nano Lett* **12**(12): 6043–6048 (2012)
- [34] Leven I, Maaravi T, Azuri I, Kronik L, Hod O. Interlayer potential for graphene/h-BN heterostructures. *J Chem Theory Comput* **12**(6): 2896–2905 (2016)
- [35] Levita G, Cavaleiro A, Molinari E, Polcar T, Righi M C. Sliding properties of MoS₂ layers: Load and interlayer orientation effects. *J Phys Chem C* **118**(25): 13809–13816 (2014)
- [36] Reguzzoni M, Fasolino A, Molinari E, Righi M C. Potential energy surface for graphene on graphene: *Ab initio* derivation, analytical description, and microscopic interpretation. *Phys Rev B* **86**(24): 245434 (2012)
- [37] Wang C Q, Chen W G, Zhang Y S, Sun Q, Jia Y. Effects of vdW interaction and electric field on friction in MoS₂. *Tribol Lett* **59**(1): 7 (2015)
- [38] Wang J J, Li J M, Li C, Cai X L, Zhu W G, Jia Y. Tuning the nanofriction between two graphene layers by external electric fields: A density functional theory study. *Tribol Lett* **61**(1): 4 (2016)
- [39] Wang L F, Ma T B, Hu Y Z, Wang H. Atomic-scale friction in graphene oxide: An interfacial interaction perspective from first-principles calculations. *Phys Rev B* **86**(12): 125436 (2012)
- [40] She H, Wang B. A geometrically nonlinear finite element model of nanomaterials with consideration of surface effects. *Finite Elem Anal Des* **45**(6–7): 463–467 (2009)
- [41] Mohammadpour E, Awang M. Nonlinear finite-element modeling of graphene and single- and multi-walled carbon nanotubes under axial tension. *Appl Phys A* **106**(3): 581–588 (2012)
- [42] Bragg W H. *An Introduction to Crystal Analysis*. London (UK): G. Bell and Sons, Ltd., 1928.
- [43] Tomlinson G A. A molecular theory of friction. *The London, Edinburgh, and Dublin Philosophical Magazine and Journal of Science* **7**(S46): 905–939 (1929)
- [44] Shinjo K, Hirano M. Dynamics of friction: Superlubric state. *Surf Sci* **283**(1–3): 473–478 (1993)
- [45] Hirano M, Shinjo K, Kaneko R, Murata Y. Anisotropy of frictional forces in muscovite mica. *Phys Rev Lett* **67**(19): 2642–2645 (1991)
- [46] Hirano M, Shinjo K, Kaneko R, Murata Y. Observation of superlubricity by scanning tunneling microscopy. *Phys Rev Lett* **78**(8): 1448–1451 (1997)
- [47] Hirano M, Shinjo K. Atomistic locking and friction. *Phys Rev B* **41**(17): 11837–11851 (1990)
- [48] Dienwiebel M, Pradeep N, Verhoeven G S, Zandbergen H W, Frenken J W M. Model experiments of superlubricity of graphite. *Surf Sci* **576**(1–3): 197–211 (2005)
- [49] Dienwiebel M, Verhoeven G S, Pradeep N, Frenken J W, Heimberg J A, Zandbergen H W. Superlubricity of graphite. *Phys Rev Lett* **92**(12): 126101 (2004)
- [50] Martin J M, Donnet C, Le Mogne T, Epicier T. Superlubricity of molybdenum disulphide. *Phys Rev B* **48**(14): 10583–10586 (1993)
- [51] Ouyang W G. New reduced models for structural superlubricity. Doctor's thesis. Beijing (China): Tsinghua University, 2016.
- [52] Pu J B, Wang L P, Xu Q J. Progress of tribology of graphene and graphene-based composite lubricating materials. *Tribology* **34**(1): 93–112 (2014)
- [53] Feng X F, Kwon S K, Park J Y, Salmeron M. Superlubric sliding of graphene nanoflakes on graphene. *ACS Nano* **7**(2): 1718–1724 (2013)
- [54] Li H, Wang J H, Gao S, Chen Q, Peng L M, Liu K H, Wei X L. Superlubricity between MoS₂ monolayers. *Adv Mater* **29**(27): 1701474 (2017)
- [55] Hod O. Superlubricity - a new perspective on an established paradigm. arXiv preprint arXiv:1204.3749, 2012.
- [56] Yang J R, Liu Z, Grey F, Xu Z P, Li X D, Liu Y L, Urbakh M, Cheng Y, Zheng Q S. Observation of high-speed microscale superlubricity in graphite. *Phys Rev Lett* **110**(25): 255504 (2013)
- [57] Filippov A E, Dienwiebel M, Frenken J W M, Klafter J, Urbakh M. Torque and twist against superlubricity. *Phys Rev Lett* **100**(4): 046102 (2008)
- [58] van Wijk M M, Dienwiebel M, Frenken J W M, Fasolino A. Superlubric to stick-slip sliding of incommensurate graphene flakes on graphite. *Phys Rev B* **88**(23): 235423 (2013)
- [59] de Wijn A S, Fusco C, Fasolino A. Stability of superlubric sliding on graphite. *Phys Rev E* **81**(4): 046105 (2010)
- [60] Liu Y L, Grey F, Zheng Q S. The high-speed sliding friction of graphene and novel routes to persistent superlubricity. *Sci Rep* **4**: (2015)
- [61] Berman D, Deshmukh S A, Sankaranarayanan S K R S, Erdemir A, Sumant A V. Macroscale superlubricity enabled by graphene nanoscroll formation. *Science* **348**(6239): 1118–1122 (2015)

- [62] Wu P, Li X M, Zhang C H, Chen X C, Lin S Y, Sun H Y, Lin C T, Zhu H W, Luo J B. Self-assembled graphene film as low friction solid lubricant in macroscale contact. *ACS Appl Mater Interfaces* **9**(25): 21554–21562 (2017)
- [63] Zhang R F, Ning Z Y, Zhang Y Y, Zheng Q S, Chen Q, Xie H H, Zhang Q, Qian W Z, Wei F. Superlubricity in centimetres-long double-walled carbon nanotubes under ambient conditions. *Nat Nanotechnol* **8**(12): 912–916 (2013)
- [64] Liu S W, Wang H P, Xu Q, Ma T B, Yu G, Zhang C H, Geng D C, Yu Z W, Zhang S G, Wang W Z, Hu Y Z, Wang H, Luo J B. Robust microscale superlubricity under high contact pressure enabled by graphene-coated microsphere. *Nat Commun* **8**: 14029 (2017)
- [65] Vu C C, Zhang S M, Urbakh M, Li Q Y, He Q C, Zheng Q S. Observation of normal-force-independent superlubricity in mesoscopic graphite contacts. *Phys Rev B* **94**(8): 081405 (2016)
- [66] Hod O. Interlayer commensurability and superlubricity in rigid layered materials. *Phys Rev B* **86**(7): 075444 (2012)
- [67] Cahangirov S, Ciraci S, Özçelik V O. Superlubricity through graphene multilayers between Ni(111) surfaces. *Phys Rev B* **87**(20): 205428 (2013)
- [68] Guo Y F, Qiu J P, Guo W L. Reduction of interfacial friction in commensurate graphene/h-BN heterostructures by surface functionalization. *Nanoscale* **8**(1): 575–580 (2016)
- [69] Leven I, Krepel D, Shemesh O, Hod O. Robust superlubricity in graphene/h-BN heterojunctions. *J Phys Chem Lett* **4**(1): 115–120 (2013)
- [70] Matsushita K, Matsukawa H, Sasaki N. Atomic scale friction between clean graphite surfaces. *Solid State Commun* **136**(1): 51–55 (2005)
- [71] Sasaki N, Kobayashi K, Tsukada M. Atomic-scale friction image of graphite in atomic-force microscopy. *Phys Rev B* **54**(3): 2138–2149 (1996)
- [72] Sasaki N, Tsukada M, Fujisawa S, Sugawara Y, Morita S, Kobayashi K. Load dependence of the frictional-force microscopy image pattern of the graphite surface. *Phys Rev B* **57**(7): 3785–3786 (1998)
- [73] Guo Y F, Guo W L, Chen C F. Modifying atomic-scale friction between two graphene sheets: A molecular-force-field study. *Phys Rev B* **76**(15): 155429 (2007)
- [74] Bonelli F, Manini N, Cadelano E, Colombo L. Atomistic simulations of the sliding friction of graphene flakes. *Eur Phys J B* **70**(4): 449–459 (2009)
- [75] Xu L, Ma T B, Hu Y Z, Wang H. Vanishing stick–slip friction in few-layer graphenes: The thickness effect. *Nanotechnology* **22**(28): 285708 (2011)
- [76] Cannara R J, Brukman M J, Cimatú K, Sumant A V, Baldelli S, Carpick R W. Nanoscale friction varied by isotopic shifting of surface vibrational frequencies. *Science* **318**(5851): 780–783 (2007)
- [77] Filletter T, McChesney J L, Bostwick A, Rotenberg E, Emtsev K V, Seyller T, Horn K, Bennewitz R. Friction and dissipation in epitaxial graphene films. *Phys Rev Lett* **102**(8): 086102 (2009)
- [78] Dong Y L. Effects of substrate roughness and electron–phonon coupling on thickness-dependent friction of graphene. *J Phys D Appl Phys* **47**(5): 055305 (2014)
- [79] Lee C G, Wei X D, Li Q Y, Carpick R, Kysar J W, Hone J. Elastic and frictional properties of graphene. *Phys Status Solidi B* **246**(11–12): 2562–2567 (2009)
- [80] Li Q Y, Lee C G, Carpick R W, Hone J. Substrate effect on thickness-dependent friction on graphene. *Phys Status Solidi B* **247**(11–12): 2909–2914 (2010)
- [81] Cho D H, Wang L, Kim J S, Lee G H, Kim E S, Lee S H, Lee S Y, Hone J, Lee C G. Effect of surface morphology on friction of graphene on various substrates. *Nanoscale* **5**(7): 3063 (2013)
- [82] Deng Z, Klimov N N, Solares S D, Li T, Xu H, Cannara R J. Nanoscale interfacial friction and adhesion on supported versus suspended monolayer and multilayer graphene. *Langmuir* **29**(1): 235–243 (2013)
- [83] Ye Z J, Tang C, Dong Y L, Martini A. Role of wrinkle height in friction variation with number of graphene layers. *J Appl Phys* **112**(11): 116102 (2012)
- [84] Smolyanitsky A, Killgore J P, Tewary V K. Effect of elastic deformation on frictional properties of few-layer graphene. *Phys Rev B* **85**(3): 035412 (2012)
- [85] Deng Z, Smolyanitsky A, Li Q Y, Feng X Q, Cannara R J. Adhesion-dependent negative friction coefficient on chemically modified graphite at the nanoscale. *Nat Mater* **11**(12): 1032–1037 (2012)
- [86] Sun X Y, Qi Y Z, Ouyang W G, Feng X Q, Li Q Y. Energy corrugation in atomic-scale friction on graphite revisited by molecular dynamics simulations. *Acta Mech Sin* **32**(4): 604–610 (2015)
- [87] Reguzzoni M, Fasolino A, Molinari E, Righi M C. Friction by shear deformations in multilayer graphene. *J Phys Chem C* **116**(39): 21104–21108 (2012)
- [88] Wu Y Y, Tsui W C, Liu T C. Experimental analysis of tribological properties of lubricating oils with nanoparticle additives. *Wear* **262**(7–8): 819–825 (2007)
- [89] Song X Y, Zheng S H, Zhang J, Li W, Chen Q, Cao B Q. Synthesis of monodispersed ZnAl₂O₄ nanoparticles and their tribology properties as lubricant additives. *Mater Res Bull* **47**(12): 4305–4310 (2012)

- [90] Chiñas-Castillo F, Spikes H A. Mechanism of action of colloidal solid dispersions. *J Tribol* **125**(3): 552–557 (2003)
- [91] Xu T, Zhao J Z, Xu K. The ball-bearing effect of diamond nanoparticles as an oil additive. *J Phys D Appl Phys* **29**(11): 2932–2937 (1996)
- [92] Rapoport L, Leshchynsky V, Lvovsky M, Nepomnyashchy O, Volovik Y, Tenne R. Mechanism of friction of fullerenes. *Ind Lubr Tribol* **54**(4): 171–176 (2002)
- [93] Hu Z S, Lai R, Lou F, Wang L G, Chen Z L, Chen G X, Dong J X. Preparation and tribological properties of nanometer magnesium borate as lubricating oil additive. *Wear* **252**(5–6): 370–374 (2002)
- [94] Kalin M, Kogovšek J, Remškar M. Mechanisms and improvements in the friction and wear behavior using MoS₂ nanotubes as potential oil additives. *Wear* **280–281**: 36–45 (2012)
- [95] Verma A, Jiang W P, Abu Safe H H, Brown W D, Malshe A P. Tribological behavior of deagglomerated active inorganic nanoparticles for advanced lubrication. *Tribol Trans* **51**(5): 673–678 (2008)
- [96] Chou R, Battez A H, Cabello J J, Viesca J L, Osorio A, Sagastume A. Tribological behavior of polyalphaolefin with the addition of nickel nanoparticles. *Tribol Int* **43**(12): 2327–2332 (2010)
- [97] Ginzburg B M, Shibaev L A, Kireenko O F, Shepelevskii A A, Baidakova M V, Sitnikova A A. Antiwear effect of fullerene C₆₀ additives to lubricating oils. *Russ J Appl Chem* **75**(8): 1330–1335 (2002)
- [98] Zhou X D, Xun F, Shi H Q, Hu Z S. Lubricating properties of Cyanex 302-modified MoS₂ microspheres in base oil 500SN. *Lubr Sci* **19**(1): 71–79 (2007)
- [99] Liu G, Li X, Qin B, Xing D, Guo Y, Fan R. Investigation of the mending effect and mechanism of copper nano-particles on a tribologically stressed surface. *Tribol Lett* **17**(4): 961–966 (2004)
- [100] Sui T Y, Song B Y, Zhang F, Yang Q X. Effect of particle size and ligand on the tribological properties of amino functionalized hairy silica nanoparticles as an additive to polyalphaolefin. *J Nanomater* **16**(1): 427 (2015)
- [101] Yan Q, Feng A C, Zhang H J, Yin Y W, Yuan J Y. Redox-switchable supramolecular polymers for responsive self-healing nanofibers in water. *Polym Chem* **4**(4): 1216–1220 (2013)
- [102] Zhang K, Li H P, Shi Q, Xu J, Zhang H T, Chen L, Li C S. Synthesis and tribological properties of Ti-doped WSe₂ nanoflakes. *Chalcogedine Lett* **12**(2): 51–57 (2015)
- [103] Liang S S. Liquid exfoliation of two-dimensional nanomaterials aiming for tribological applications. Doctor's thesis. Beijing (China): Beihang University, 2015.
- [104] Hu K H, Huang F, Hu X G, Xu Y F, Zhou Y Q. Synergistic effect of nano-MoS₂ and anatase nano-TiO₂ on the lubrication properties of MoS₂/TiO₂ nano-clusters. *Tribol Lett* **43**(1): 77–87 (2011)
- [105] Su Y, Gong L, Chen D D. An investigation on tribological properties and lubrication mechanism of graphite nanoparticles as vegetable based oil additive. *J Nanomater* **16**(1): 203 (2015)
- [106] Xiao H P, Liu S H. 2D nanomaterials as lubricant additive: A review. *Mater Des* **135**: 319–332 (2017)
- [107] Gulzar M, Masjuki H H, Kalam M A, Varman M, Zulkifli N W M, Mufti R A, Zahid R. Tribological performance of nanoparticles as lubricating oil additives. *J Nanopart Res* **18**(8): 223 (2016)
- [108] Meng Y, Su F H, Chen Y Z. Supercritical fluid synthesis and tribological applications of silver nanoparticle-decorated graphene in engine oil nanofluid. *Sci Rep* **6**: 31246 (2016)
- [109] Dai W, Kheireddin B, Gao H, Kan Y W, Clearfield A, Liang H. Formation of anti-wear tribofilms via α -ZrP nanoplatelet as lubricant additives. *Lubricants* **4**(3): 28 (2016)
- [110] Berman D, Erdemir A, Sumant A V. Few layer graphene to reduce wear and friction on sliding steel surfaces. *Carbon* **54**: 454–459 (2013)
- [111] Berman D, Erdemir A, Sumant A V. Reduced wear and friction enabled by graphene layers on sliding steel surfaces in dry nitrogen. *Carbon* **59**: 167–175 (2013)
- [112] Zhao L, Cai Z B, Zhang Z C, Zhang X, Lin Y W, Peng J F, Zhu M H. Tribological properties of graphene as effective lubricant additive in oil on textured bronze surface. *Chin J Mater Res* **30**(1): 57–62 (2016)
- [113] Senatore A, D'Agostino V, Petrone V, Ciambelli P, Sarno M. Graphene oxide nanosheets as effective friction modifier for oil lubricant: Materials, methods, and tribological results. *ISRN Tribol* **2013**: 425809 (2013)
- [114] Zhang W, Zhu H W. Graphene enhanced lubricant oil. *Chin J Nat* **38**(2): 94–96 (2016)
- [115] Zhang W, Zhou M, Zhu H W, Tian Y, Wang K L, Wei J Q, Ji F, Li X, Li Z, Zhang P, Wu D H. Tribological properties of oleic acid-modified graphene as lubricant oil additives. *J Phys D Appl Phys* **44**(20): 205303 (2011)
- [116] Lin J S, Wang L W, Chen G H. Modification of graphene platelets and their tribological properties as a lubricant additive. *Tribol Lett* **41**(1): 209–215 (2011)
- [117] Zheng S Z, Zhou Q, Yang S R, Yang Z G, Wang J Q. Preparation and tribological properties of fluorinated graphene nanosheets as additive in lubricating oil. *Tribology* **37**(3): 402–408 (2017)

- [118] Dou X, Koltonow A R, He X L, Jang H D, Wang Q, Chung Y W, Huang J X. Self-dispersed crumpled graphene balls in oil for friction and wear reduction. *Proc Natl Acad Sci USA* **113**(6): 1528–1533 (2016)
- [119] Song H J, Li N. Frictional behavior of oxide graphene nanosheets as water-base lubricant additive. *Appl Phys A* **105**(4): 827–832 (2011)
- [120] Cho D H, Kim J S, Kwon S H, Lee C G, Lee Y Z. Evaluation of hexagonal boron nitride nano-sheets as a lubricant additive in water. *Wear* **302**(1–2): 981–986 (2013)
- [121] He X L, Xiao H P, Choi H, Diaz A, Mosby B, Clearfield A, Liang H. α -Zirconium phosphate nanoplatelets as lubricant additives. *Colloid Surf A* **452**: 32–38 (2014)
- [122] Zhao B H. Sputtering technique of MoS₂ solid lubricant. *Bearing* (2): 31–33 (2002)
- [123] Renevier N M, Oosterling H, König U, Dautzenberg H, Kim B J, Geppert L, Koopmans F G M, Leopold J. Performance and limitations of MoS₂/Ti composite coated inserts. *Surf Coat Technol* **172**(1): 13–23 (2003)
- [124] Renevier N M, Fox V C, Teer D G, Hampshire J. Coating characteristics and tribological properties of sputter-deposited MoS₂/metal composite coatings deposited by closed field unbalanced magnetron sputter ion plating. *Surf Coat Technol* **127**(1): 24–37 (2000)
- [125] Ma G Z, Xu B S, Wang H D, Si H J. State of research on space solid lubrication materials. *Mater Rev* **24**(1): 68–71 (2010)
- [126] Nian J Y, Chen L W, Guo Z G, Liu W M. Computational investigation of the lubrication behaviors of dioxides and disulfides of molybdenum and tungsten in vacuum. *Friction* **5**(1): 23–31 (2017)
- [127] Cheng Y Z. Preparation and tribological properties of space lubricating grease containing MoS₂ nanoparticles. Master's thesis. Hefei (China): Hefei University of Technology, 2012.
- [128] Luo X Y, Tang Z J, Wang D J, Chen M H. Improving antiwear properties of aviation lubricant with in situ synthesis' nano MoS₂ particle/PR. *Lubr Eng* (5): 57–58 (2003)
- [129] Song H, Ji L, Li H X, Wang J Q, Liu X H, Zhou H D, Chen J M. Self-forming oriented layer slip and macroscale super-low friction of graphene. *Appl Phys Lett* **110**(7): 073101 (2017)
- [130] Lee J H, Kim S H, Cho D H, Kim S C, Baek S G, Lee J G, Kang J M, Choi J B, Seok C S, Kim M K, Koo J C, Lim B S. Tribological properties of chemical vapor deposited graphene coating layer. *Korean J Met Mater* **50**(3): 206–211 (2012)
- [131] Watanabe S, Noshiro J, Miyake S. Tribological characteristics of WS₂/MoS₂ solid lubricating multilayer films. *Surf Coat Technol* **183**(2–3): 347–351 (2004)
- [132] Kim K S, Lee H J, Lee C G, Lee S K, Jang H, Ahn J H, Kim J H, Lee H J. Chemical vapor deposition-grown graphene: The thinnest solid lubricant. *ACS Nano* **5**(6): 5107–5114 (2011)
- [133] Sun H Y, Li X M, Li Y C, Chen G X, Liu Z D, Alam F E, Dai D, Li L, Tao L, Xu J B, Fang Y, Li X S, Zhao P, Jiang N, Chen D, Lin C T. High-quality monolithic graphene films via laterally stitched growth and structural repair of isolated flakes for transparent electronics. *Chem Mater* **29**(18): 7808–7815 (2017)
- [134] Li L, Li X M, Du M D, Guo Y C, Li Y C, Li H B, Yang Y, Alam F E, Lin C T, Fang Y. Solid-phase coalescence of electrochemically exfoliated graphene flakes into a continuous film on copper. *Chem Mater* **28**(10): 3360–3366 (2016)
- [135] Hirano M. Atomistics of superlubricity. *Friction* **2**(2): 95–105 (2014)
- [136] Zheng Q S, Liu Z. Experimental advances in superlubricity. *Friction* **2**(2): 182–192 (2014)
- [137] Tan C L, Zhang H. Wet-chemical synthesis and applications of non-layer structured two-dimensional nanomaterials. *Nat Commun* **6**: 7873 (2015)
- [138] Wang F, Wang Z X, Shifa T A, Wen Y, Wang F M, Zhan X Y, Wang Q S, Xu K, Huang Y, Yin L, Jiang C, He J. Two-dimensional non-layered materials: Synthesis, properties and applications. *Adv Funct Mater* **27**(19): 1603254 (2017)



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