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Review Article

Structural insights into 2D materials for tribology

Lei Liu[1,2*], Pengxu Yang[1], Mengyu Wang[1], Linjian Shangguan [1,*], Han Peng [1], Zhengquan Jiang[2], Chenhua Liu[4], Yuhong Liu[3,*]

[1] *School of Mechanical Engineering, North China University of Water Resources and Electric Power, Zhengzhou 450045, China*

[2] *Institute of Special Friction and Lubricating Materials, North China University of Water Resources and Electric Power, Zhengzhou 450045, China*

[3] *State Key Laboratory of Tribology in Advanced Equipment, Tsinghua University, Beijing 100084, China*

[4] *School of Mechanical Engineering, North University of China, Taiyuan 030051, China*

* Corresponding author: L. Liu, E-mail: liu-l18@tsinghua.org.cn; L. Shangguan, E-mail: shangguanlinjian@ncwu.edu.cn; Y. Liu, E-mail: liuyuhong@tsinghua.edu.cn

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Abstract: Owing to their excellent mechanical properties, two-dimensional (2D) materials show substantial promise for application in tribology. This has prompted a proliferation of novel 2D structures in numerous studies. However, there is a lack of understanding regarding the intrinsic

relationship between their structure and lubrication performance, which presents a challenge to the systematic design of high-performance 2D lubricating materials. Therefore, this paper first classifies 2D lubricating materials into four categories based on their structural characteristics: inorganic, organic, carbon-based, and hybrid structures. Subsequently, it systematically reviews key research advances for each structural type within the field of tribology. This review encompasses fundamental studies on intrinsic lubrication ability—primarily investigated through microscopic experiments and theoretical simulations—as well as applied research exploring their use as lubricant additives (both oil- and water-based), lubricant coatings, and reinforcing phases in composite materials. Furthermore, the lubrication mechanisms of diverse 2D structures across various lubrication regimes have been systematically summarized, and strategies for enhancing performance via structural modification are also discussed. Finally, the current challenges and future research directions for 2D lubricating materials are critically analyzed.

Keywords: 2D Materials; Tribology; Structural Perspective; Lubrication Mechanism; Structure-Performance Relationship

1 Introduction

According to statistics, the energy consumed by friction accounts for more than a quarter of the total energy supplied from fossil fuels each year[1]. Moreover, the issue of friction and wear at the microscale is particularly worthy of attention[2-5], as it hinders the development and application of devices smaller than a millimeter. However, the emergence of superlubricity technology will bring unprecedented progress to the field of microsystems[6,7]. The following cases illustrate the potential of superlubricity technology in micro/nano-electromechanical systems (M/NEMS): it significantly improves the performance and durability of micro-motors[8], making the design of mechanical hands of humanoid robot possible; in addition, this technology can be applied to micro-generators[9] to meet

the requirements about long-term power supply and maintenance-free of ubiquitously distributed micro-sensors in smart grids and promote an increase in power generation; and, the superlubricity technology endows the Radio Frequency (RF) switches[10] with features of miniaturization, low loss, high power output and long service life that will drive the advancement of wireless communication terminal devices.

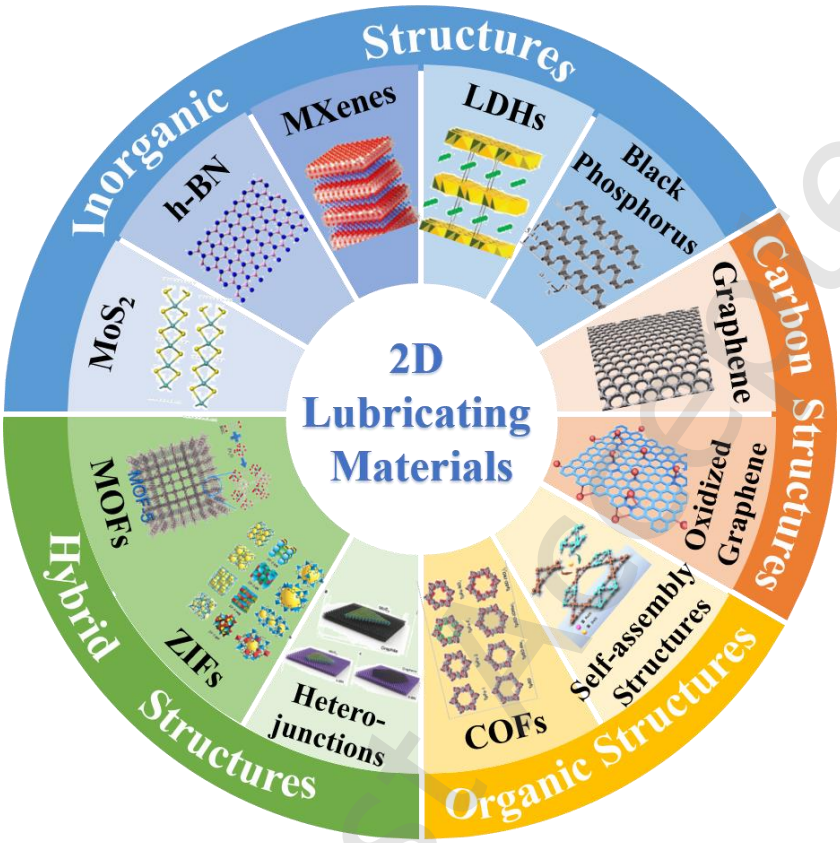

Fig. 1 Overview of 2D lubricating materials with specific structural classification.

The reported microscopic friction mechanisms[11] have verified the great potential of two-dimensional (2D) materials[12] in the field of superlubricity[13]. The layer structure of 2D materials, which possess strong bonds in laminates and weak interactions between layers, gives them excellent mechanical properties[14] and makes it easy to form the shearable tribofilms/transfer layers[15]. Moreover, there is an ideal physical model for the interlayer sliding process of 2D materials, known as structural superlubricity[16,17], where the contacting crystal planes are in an incommensurable state, and the sliding friction force disappears. With the vigorous development of superlubricity technology,

there has also been an explosion of superlubricating materials[18-20]. Although there have been several reviews on 2D materials in the field of lubrication[21-25], including the summaries of application forms[26-29], performance regulation[14] and lubrication mechanisms[11], the analysis of their inherent lubrication capabilities from a structural perspective is lacking that has hindered the formation of a clear structure-performance relationship among tribologists. We have summarized the 2D materials that have attracted much attention in the field of lubrication, and classified them into the following four categories based on the constituent units and the forms of core chemical bond in their laminate structures: inorganic materials, organic materials, carbon materials, and hybrid materials (Figure 1).

The primary chemical bonds in the single-layer structure of 2D inorganic materials are typically covalent bonds, though ionic interactions may also play a role. These materials are composed of various constituent units, including metal oxides, sulfides, carbides, nitrides, halides, and hydroxides, as well as non-metal elements, oxides, carbides, and nitrides; 2D organic materials consist exclusively of light elements such as B, C, N, O, Si, and H, with carbon being the primary component. Their skeletal structure is formed by covalent bonds, hydrogen bonds, or van der Waals (VDW) forces between organic molecules; 2D carbon materials share some similarities with both inorganic and organic materials in structure, but due to their significant role in the field of lubrication, they have been classified separately. Their core chemical bond is C-C covalent bonds, and only the crystal defects and edges can be terminated by H, F and O elements or OH groups; In recent years, 2D hybrid materials have emerged as a new force in the field of lubrication. As the name implies, the hybrid structure is composed of two or more components with distinct physicochemical properties. For example, the inorganic-organic hybrid structure is formed by the periodic arrangements of inorganic and organic components, with their core chemical bond being coordination bonds and covalent bonds, and metal-organic frameworks (MOFs) is the representative structure.

In the following subsections, we will summarize the important research status of specific 2D structures of four types of lubricating materials in the field of tribology. For each substructure, we will first introduce its structural characteristics, and then successively summarize the research progress in the fields of intrinsic lubrication ability, lubricant additives, lubricant coatings or reinforcement phases, where the intrinsic lubrication ability includes the lubrication mechanisms and performance regulation capability investigated by the nanoscale/microscale experiments and theoretical simulations[30]. Given that the preparation process of 2D materials and the experimental investigation into their intrinsic lubrication properties predominantly rely on the innovative insights and creative approaches of researchers, this paper initially focuses on summarizing the synthesis methodologies of 2D materials and the characterization techniques employed in nanotribology before proceeding to provide a comprehensive overview of the advancements in the field of tribology.

2 Synthetic Methodology of 2D Materials

The preparation of 2D materials primarily falls into two categories: "top-down" and "bottom-up" approaches[31], each encompassing multiple techniques tailored for specific material types and applications (Figure 2). Below is a structured overview of these methods, integrating principles and illustrative case studies.

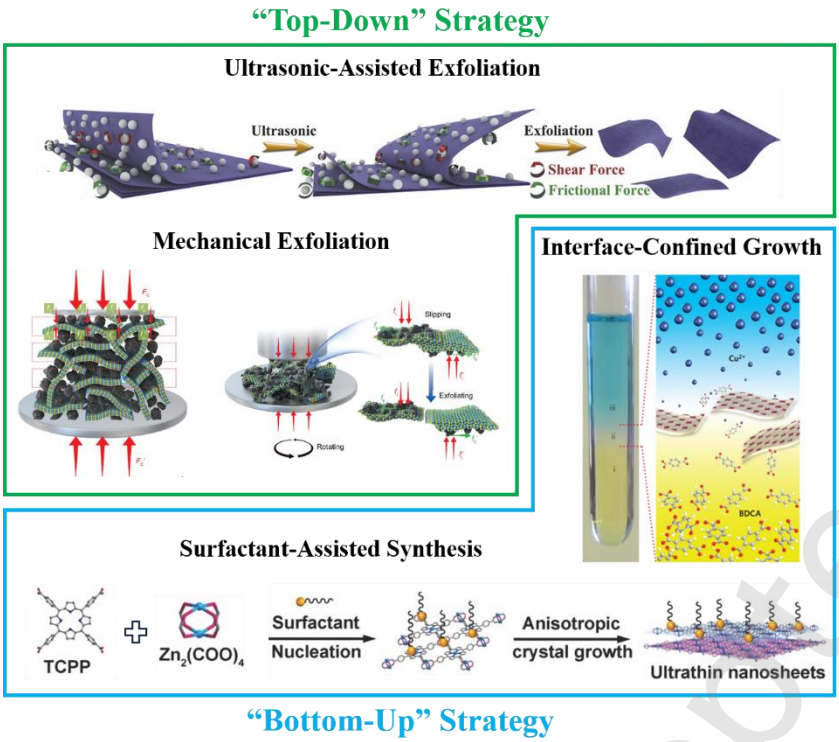

Fig. 2 Methodology for the synthesis of 2D materials.

2.1 Top-Down Methods

These methods begin with bulk materials and exfoliate 2D materials through physical processes, making them particularly suited for layered materials such as graphite and molybdenum disulfide (MoS_2). The core idea is to mechanically or sonically disrupt the weak van der Waals forces between layers, enabling the isolation of single- or few-layer sheets.

A prominent technique within this category is mechanical exfoliation, which relies on physical force to peel away layers. The Scotch Tape Method exemplifies this approach, where repeated peeling of bulk material using adhesive tape yields high-quality nanosheets—a breakthrough demonstrated by Novoselov and colleagues[32] in 2004, who produced graphene flakes up to 100 micrometers in size. Another variant is ball milling, which employs grinding media to generate collision and shear forces for large-scale production; however, this method may introduce structural defects due to the harsh mechanical environment. In 2020, Liu et al.[33] introduced a novel grinding exfoliation technique that employs micro-particles as force intermediates to convert applied compressive forces into numerous

small shear forces, enabling highly efficient exfoliation of layered materials. Termed intermediate-assisted grinding exfoliation (iMAGE), this method facilitates large-scale production of diverse 2D materials. For instance, bulk hexagonal boron nitride (h-BN) was exfoliated into 2D h-BN with large flake sizes, high quality, and structural integrity, achieving an impressive exfoliation yield of 67%, a production rate of 0.3 g h^{-1}, and low energy consumption of 3.01×10^{6} J g^{-1}. Beyond h-BN, iMAGE has demonstrated universality by exfoliating graphite, black phosphorus, transition metal dichalcogenides, and metal oxides. Notably, the method was successfully applied to molybdenite concentrate, a naturally abundant and low-cost mineral, for large-scale production of 2D MoS_2 flakes. This work highlights iMAGE's potential to produce large quantities of various 2D materials, marking a significant step toward their commercial application.

Complementing mechanical methods is ultrasonic-assisted exfoliation, which utilizes the cavitation and shear forces generated by ultrasound in a liquid medium to separate layers. This approach offers higher production yields but requires careful optimization of parameters such as power and duration to prevent material damage, ensuring the integrity of the exfoliated sheets. In 2020, Wang et al.[34] introduced an innovative and straightforward "ultrasonic-ball milling" strategy, which effectively produces high-quality, large-sized ultrathin 2D materials with intact lattice structures by incorporating moderate sapphire (Al_2O_3) abrasives in a liquid-phase system. This method synergistically harnesses shear and friction forces, enabling the scalable production of ultrathin nanosheets of h-BN, graphene, MoS_2, WS_2, and BCN with dimensions ranging from 1–20 μm, thicknesses approaching 1–3 nm, and yields exceeding 20%. The work underscores the versatility of hybrid mechanical-ultrasonic techniques in advancing the commercial application of 2D materials.

2.2 Bottom-Up Methods

In contrast to top-down approaches, bottom-up methods construct 2D materials by assembling atoms or molecules from precursors, providing greater flexibility to synthesize both layered and non-layered

materials. These techniques enable precise control over material properties and are widely used for innovative applications.

A key strategy in this domain is interface-confined growth, which directs precursor reactions at specific interfaces to form 2D structures. The solid-gas interface method involves growing materials through vapor-phase reactions, such as chemical vapor deposition (CVD)[35], on solid substrates. Alternatively, the solid-liquid interface approach induces 2D growth via substrate-mediated reactions, including molecular self-assembly[36]. For ordered materials like metal-organic frameworks (MOFs) and covalent organic frameworks (COFs), the liquid-liquid interface technique guides growth at miscible/immiscible liquid boundaries, leveraging interfacial interactions to achieve uniformity. In 2018, Gascon et al.[37] introduced an innovative miscible liquid/liquid interface-confined synthesis strategy for producing freestanding MOF lamellae with micrometre-scale lateral dimensions and nanometre-scale thickness. This method demonstrates excellent universality, enabling the successful synthesis of two-dimensional MOFs using copper, cobalt, or zinc ions paired with benzoic acid or naphthalene carboxylic acid ligands. The technique involves sequentially adding three solutions to a test tube to form three distinct layers: a bottom layer containing the dissolved ligand, an upper layer with the dissolved metal salt, and an intermediate solvent layer. The synthesis medium comprises three vertically arranged liquid layers, each consisting of a carefully controlled mixture of N,N-dimethyl formamide (DMF) and a suitable miscible co-solvent in specific ratios, determined by their differing densities. This method relies on the diffusion-mediated modulation of the MOF growth kinetics.

Another important method is surfactant-assisted synthesis, where surfactants stabilize precursors or reaction intermediates, effectively suppressing unwanted three-dimensional growth and promoting two-dimensional formation. This technique is particularly valuable for producing colloidal nanosheets or hybrid organic-inorganic materials, offering pathways to tailor material functionality for diverse applications. In 2015, Zhang et al.[38] introduced a surfactant-assisted synthetic approach to fabricate uniform ultrathin 2D MOF nanosheets with thicknesses below 10 nm. The method employs

polyvinylpyrrolidone (PVP) as the surfactant, and the resulting MOFs feature a paddlewheel structural motif, which are formed by copper (Cu), cadmium (Cd), cobalt (Co), or zinc (Zn) ions coordinated with the tetrakis(4-carboxyphenyl)porphyrin (TCPP) ligand. In this technique, surfactant molecules selectively adsorb onto the surface of the MOFs, playing a pivotal role in controlling the growth of MOF crystals. This selective adsorption promotes anisotropic growth of the MOFs, ultimately leading to the formation of ultrathin MOF nanosheets. Beyond PVP, anionic (e.g., SDS) and cationic surfactants (e.g., CTAB) can similarly induce 2D MOF growth by selectively templating crystal surfaces[39], demonstrating the versatility of surfactant-assisted synthesis in controlling MOF dimensionality.

2.3 Comparison and Challenges

While top-down methods are celebrated for their operational simplicity and high yield in cases like ball milling, they often struggle with controlling crystal quality and size uniformity, which can limit performance in advanced applications. Bottom-up methods, on the other hand, provide strong design flexibility and precise control over material properties through adjustable reaction conditions such as temperature and concentration. However, these advantages come with scalability challenges, as large-scale production demands meticulous optimization and often faces higher costs. Understanding these trade-offs is essential for selecting the appropriate method based on specific material requirements and application goals.

3 Nanoscale tribological characterization techniques

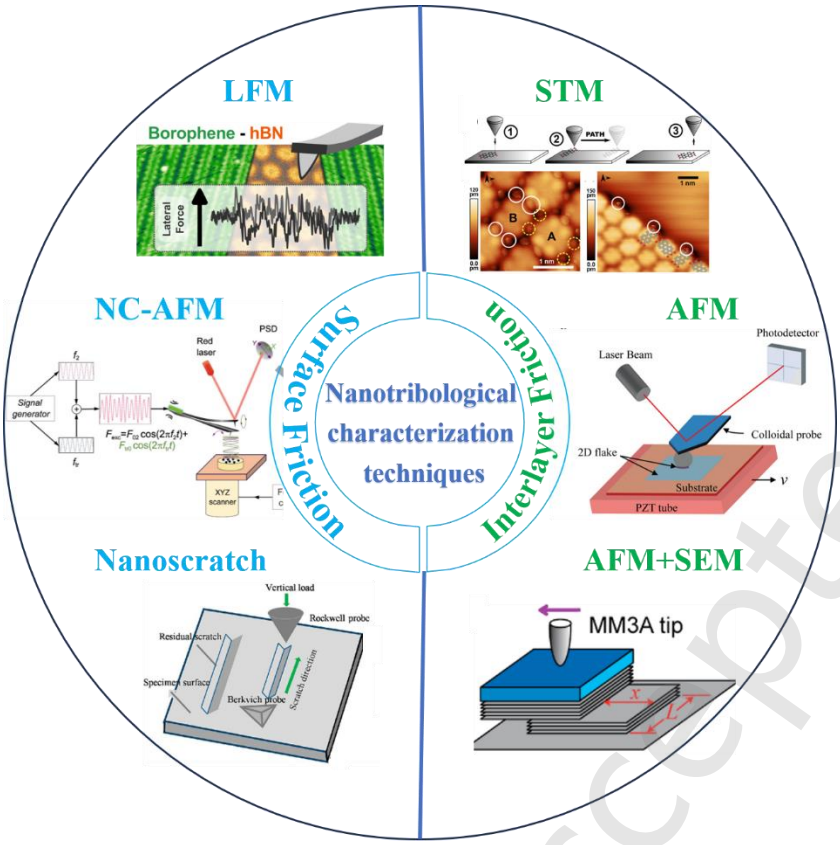

Fig. 3 Microscale tribological characterization: surface and interlayer friction analysis.

2D materials hold great promise for applications in lubrication and superlubrication. However, significant differences exist in their intrinsic lubrication capabilities and friction energy dissipation mechanisms, which are highly dependent on their structural properties. To address this, comprehensive research combining microscale experiments and atomic-scale simulations is essential. At the experimental level, this requires advanced characterization equipment and innovative testing methodologies. This section reviews the micro-friction testing techniques for 2D materials, categorizing the discussion into surface friction testing and interlayer friction testing approaches (Figure 3).

3.1 Surface friction characterization techniques

2D materials possess atomically smooth surfaces, which inherently minimize friction contributions from surface roughness. This unique characteristic makes them ideal model systems for fundamental

studies of surface friction. Consequently, characterizing their tribological properties requires testing techniques with nanoscale resolution, such as atomic force microscopy (AFM) or scanning tunneling microscopy (STM), to accurately probe their intrinsic interfacial interactions. The Lateral Force Microscope (LFM) is a significant functional extension of the Atomic Force Microscope (AFM). Building upon the fundamental principles of AFM, it is specifically designed to investigate the frictional properties of material surfaces with high precision. The key advantages of AFM in measuring microscopic friction stem from its unique combination of nanoscale resolution, multi-environment adaptability, and powerful functional integration. By seamlessly coupling with external fields like electric fields and temperature, AFM not only enables precise characterization of friction behavior at the atomic level but also provides deep insights into the underlying physical and chemical mechanisms. This makes AFM an indispensable tool in the field of nanotribology research. Non-contact atomic force microscopy (NC-AFM) indirectly measures the interaction force gradient between the tip and the sample surface by precisely monitoring minute shifts in the cantilever's resonance frequency or oscillation amplitude. This enables nanoscale characterization of friction, with the core principle relying on the synergistic interplay between highly sensitive sensors and advanced feedback systems. Nanoscratch testing quantitatively evaluates micro-friction by capturing tangential forces in real-time with a force sensor and correlating them with post-scratch morphology analysis. This method, based on controlled mechanical loading and multi-parameter monitoring, provides a reliable approach for studying the frictional performance of material surfaces.

3.1.1 AFM (LFM)

The Lateral Force Microscope (LFM) represents a specialized functional extension of Atomic Force Microscopy (AFM), designed to investigate surface friction properties with exceptional precision. While conventional AFM imaging relies on detecting vertical cantilever deflections to map surface topography, LFM uniquely captures torsional bending of the microcantilever during lateral scanning.

This critical distinction allows LFM to resolve friction variations between different material components that remain indiscernible in standard height-based topographic images.

The technical foundation of LFM lies in its ability to measure minute differences in laser beam intensity across the four quadrants of the photodetector. In standard contact-mode AFM, these intensity differences correspond to vertical deflections used for surface mapping. However, in LFM operation, when the probe scans perpendicular to the cantilever axis, the same intensity variations reflect torsional forces generated by lateral friction interactions, thereby providing quantitative friction data at the nanoscale.

This technique offers particular value in analyzing multi-component material surfaces, where it can reveal critical friction contrasts invisible in conventional topographical images. The application spectrum spans critical fields including materials science and biomedicine, where understanding surface friction is essential for developing advanced materials and biological interfaces.

LFM's technical advantages are particularly noteworthy:

1) Nanometer Resolution and Non-Destructive Measurement: AFM's atomic-scale interaction detection (with vertical resolution reaching 0.01 nm) enables LFM to observe friction phenomena without sample damage.

2) Multi-Environment Compatibility: LFM operates effectively across diverse conditions - from ambient air to vacuum and liquid environments - making it ideal for studying biological samples like live cells and proteins under realistic conditions.

3) Advanced Function Integration: The modular design of AFM allows seamless integration of additional measurement capabilities, including:

①Electric/Current Field Studies: Using conductive probes or Kelvin Probe Force Microscopy to simultaneously measure surface potential and friction in conductive materials.

②Thermal Analysis: Integrated heating modules enable observation of friction coefficient changes with temperature, crucial for polymer and lubricant research.

③Multi-parameter Characterization: Beyond friction, LFM can simultaneously measure mechanical properties such as elastic modulus and adhesion, providing comprehensive material characterization.

These combined capabilities establish LFM as an indispensable tool in nanotribology, offering unparalleled insights into surface friction phenomena at the molecular level.

In 2025, Li et al.[40] conducted a comprehensive study on the synergistic lubrication between alkane molecules with varying chain lengths and graphite, employing a multi-technique approach that included AFM, scanning tunneling microscopy (STM), and molecular dynamics (MD) simulations. Their investigation revealed that superlubricity was achieved in a hexadecane environment, exhibiting an ultralow friction coefficient of 0.001—a reduction exceeding 80% compared to dodecane ($\mu = 0.006$) and nonane ($\mu = 0.009$) environments. This remarkable performance was attributed to the structural superlubricity mechanism, wherein long-chain alkane molecules exhibited ordered alignment along the graphite surface's zigzag direction, facilitating the formation of stable confined molecular layers. Additionally, the boundary slips of these molecules on the graphite surface, characterized by low energy barriers, played a crucial role in achieving ultralow friction. This work highlights the critical influence of molecular chain length and surface interactions in realizing high-performance lubrication systems.

3.1.2 NC-AFM

Non-contact atomic force microscopy (NC-AFM) measures friction by detecting the weak interaction forces between the probe and the sample surface without direct contact, relying instead on frequency modulation (FM) or amplitude modulation (AM) techniques. In the frequency modulation mode, which is the mainstream technology, the qPlus-type NC-AFM operates in an ultra-high vacuum environment. The probe cantilever vibrates at its free resonance frequency (f_0), and as the tip approaches the sample, the interaction force gradient (including van der Waals and electrostatic forces) alters the cantilever's resonance frequency (Δf). By measuring the relationship between the frequency

shift (Δf) and the tip height, the surface force distribution, including friction, is indirectly reflected. Friction manifests as a change in the force gradient, causing a frequency shift, and the frequency signal is precisely demodulated using a phase-locked loop (PLL) circuit. In the amplitude modulation mode, the cantilever vibrates at a constant amplitude, and the attractive force between the tip and the sample reduces the oscillation amplitude. A feedback system maintains the set amplitude, while the Z-axis scanner's displacement changes are recorded to infer the surface force distribution, including friction. This mode avoids tip damage and sample contamination, making it ideal for fragile surfaces like biological samples and soft materials.

The friction measurement principle in NC-AFM is indirect, achieved through force gradient analysis (where frequency or amplitude changes reflect the interaction force gradient, with friction appearing as local anomalies) and lateral force detection (where periodic changes in frequency or amplitude during lateral scanning reveal surface friction characteristics). The technical advantages of NC-AFM for friction measurement include atomic-level resolution, wide applicability to conductors, insulators, and biological samples, and non-destructive operation, preventing tip wear and sample damage common in contact AFM.

In 2025, Hinaut et al.[41] conducted a comparative experimental study on the frictional properties of borophene and h-BN using a borophene/h-BN lateral heterostructure. The authors systematically examined the friction between a sliding tip and two distinct systems: (i) the weakly corrugated X_6-borophene layer on Ir(111) and (ii) the h-BN/Ir(111) superlattice structures featuring strongly corrugated Moiré reconstruction. Through a combination of experimental measurements and Prandtl-Tomlinson (PT) model calculations, the study confirmed the excellent superlubricity performance of borophene, while also establishing h-BN as a low-friction material with comparatively higher friction. Ab initio calculations revealed that the superior frictional properties of X_6-borophene over h-BN can be attributed to weaker tip-surface interactions. Furthermore, the authors employed NC-AFM techniques to assess the structural and electrical properties of both materials, investigating energy

dissipation mechanisms under an oscillating tip. The results demonstrated lower energy dissipation in borophene compared to h-BN, suggesting a flatter and more rigid borophene layer in contrast to the more deformable Moiré structure of h-BN. This conclusion was further supported by the calculated dissipation ratio (Dr), which revealed values of $Dr_B \simeq 3$ for borophene and $Dr_{h\text{-}BN} \simeq 5.4$ for h-BN, indicating a larger variation in dissipation across the h-BN surface and consistent with its greater deformability.

3.1.3 Nanoscratch

The nano-scratch test indirectly measures microscopic friction by precisely controlling the probe-sample interaction. Its core principle involves quantifying friction behavior through real-time monitoring of the tangential force generated during scratching. The implementation methodology includes three key aspects: (1) Force sensor detection - A diamond probe (e.g., Berkovich or conical indenter) applies a vertical load while moving horizontally, with friction converted into measurable tangential forces by a high-sensitivity force sensor; (2) Friction coefficient calculation - Obtained as the ratio of tangential to normal force, reflecting material surface characteristics; (3) Morphology-force correlation - Post-scratch surface analysis (via optical microscopy or 3D profilometry) reveals relationships between friction forces and material damage mechanisms.

This technique offers several technical advantages: high-precision force sensing (down to 50 μN resolution) for nanoscale measurements; synchronized multi-parameter recording (normal load, tangential force, displacement) for comprehensive mechanical data; and broad applicability across coatings, films, and biological materials, particularly for evaluating bonding strength, wear resistance, and critical failure loads.[42]

Compared to other techniques, while AFM directly measures nanoscale friction via cantilever torsion but has limited test range, nano-scratch testing better simulates macroscopic friction behavior and quantifies coating adhesion. Similarly, though nano-indentation primarily measures hardness and elastic modulus, scratch testing focuses specifically on friction and damage mechanisms.

In 2024, Paul et al.[43] investigated frictional resistance and delamination mechanisms in 2D tungsten diselenide (WSe_2) at 10 μm length scales and depths of 100–500 nm using integrated AFM, high-load nanoscratch testing, and in-situ scanning electron microscopy (SEM). AFM revealed a heterogeneous distribution of frictional resistance in single-layer WSe_2, attributed to surface ripples, with the mean friction force increasing from 8.7 to 79.1 nN as the applied load rose from 20 to 80 nN. High-load in-situ nanoscratch tests elucidated the role of individual layers in multilayer delamination, observing stick-slip motion where friction forces increased incrementally with each slip—manifested as a rising slope in lateral force curves (from 10.9 to 13.0×10^3 Nm^{-1}) with scratch depth. Post-delamination, cyclic layer fracture occurred, with the puckering effect causing adhesion to the nanoscratch probe, elevating the lateral force peak from 89.3 to 205.6 μN. This study establishes a critical link between single-layer friction and multilayer delamination, offering insights for designing low-energy-loss, high-performance semiconductor devices.

3.2 Interfacial friction characterization techniques

The key to measuring the interlayer frictional properties of 2D materials lies in constructing a well-controlled homojunction or heterojunction of 2D materials, enabling precise shear sliding along the crystallographic plane direction while conveniently adjusting the applied normal load and sliding velocity. For interlayer adhesion characterization, it is essential to firmly bond the 2D materials to a friction pair surface capable of normal motion and equipped with a force sensor. Current state-of-the-art interlayer friction testing techniques for 2D materials primarily encompass three categories: (1) AFM-based methods, (2) integrated approaches combining AFM with complementary techniques (e.g., Scanning Electron Microscope (SEM)), and (3) scanning tunneling microscopy (STM)-based techniques. These advanced methodologies allow for atomic-scale resolution and quantitative analysis of interfacial friction and adhesion phenomena in 2D material systems.

3.2.1 AFM-based methods

The techniques for testing the interfacial friction of 2D materials using AFM have evolved into a rich and diverse set of methods. These techniques originated from AFM-based nanomanipulation technologies[44], which were initially limited in their ability to perform comprehensive interlayer adhesion tests. To overcome this limitation, researchers developed advanced approaches involving the functionalization of AFM tips[45] or microsphere probes[46,47] with 2D materials, enabling more precise and controlled measurements of interfacial friction and adhesion properties. This progression has significantly expanded the capabilities of AFM-based tribological characterization for 2D material systems.

In 2024, Qi et al.[48] employed an innovative method of wrapping 2D materials around silicon spheres to investigate interlayer friction and adhesion effects in Penta-$PdSe_2$-based van der Waals heterostructures. Using a micromanipulator integrated into the 2D material transfer system, a minimal amount of epoxy resin adhesive was applied to the tip-less end of a cantilever, enabling the meticulous attachment of an SiO_2 sphere to form a microsphere probe. Subsequently, an entire 2D material sheet pre-deposited on polydimethylsiloxane (PDMS) was introduced onto the microsphere probe, with a small additional amount of adhesive applied to the top of the silica sphere to ensure robust contact. The results revealed that graphene/MoS_2 and graphene/$PdSe_2$ heterostructure interfaces exhibited extremely low friction coefficients ($\approx 10^{-3}$), while the MoS_2/$PdSe_2$ interfaces showed higher friction coefficients (≈0.02). This disparity was attributed to significant interfacial interactions driven by interlayer charge transfer, closely linked to the ionic nature of the 2D material crystals. These findings highlight the critical role of material composition in determining friction behavior, offering insights for designing low-friction, high-performance electronic devices.

3.2.2 Integrated approaches combining AFM with complementary techniques

To meet the growing demand for test accuracy and a deeper understanding of interlayer friction characteristics, advanced characterization techniques have been progressively integrated with AFM-based interlayer friction testing. This includes in-situ observation of the interlayer sliding process using

optical microscope, scanning electron microscopy (SEM) and precise assembly of homojunctions/heterojunctions through microscale robotic arms, enabling atomic-scale manipulation and real-time monitoring of interfacial interactions. These integrated approaches enhance the precision and comprehensiveness of friction measurements in 2D material systems.

Investigating the interlayer friction of both graphite homojunctions and heterojunctions through the assembly of graphite islands represents a typical application of this technology. Zheng et al.[49] innovatively developed the graphite island assembly technique, which involves growing a silicon dioxide (SiO_2) film on the freshly cleaved surface of highly oriented pyrolytic graphite (HOPG) using plasma-enhanced chemical vapor deposition (PECVD), followed by electron beam lithography and reactive ion etching to create HOPG islands. By shearing out a top graphite/SiO_2 flake from the island using an AFM tip, the authors observed the self-retraction of the sheared flake upon tip release. In situ manipulation of a graphite/SiO_2 flake within a SEM revealed locked-up states, with angles between neighboring locked-up orientations close to 60°. This graphite island assembly technology also enabled the discovery of robust structural superlubricity in heterojunctions between graphite and hexagonal boron nitride (h-BN)[50], further demonstrating the potential of this approach for studying and engineering low-friction interfaces in 2D materials. In 2017, Liu et al.[51] advanced interlayer friction testing by integrating a SEM with an AFM and incorporating a silicon nanowire force sensor into the system, enabling precise measurements of friction in monolayer molybdenum disulfide (MoS_2). Their setup involved a single Si nanowire clamped to a tungsten (W) tip, which could move in three independent directions with nanometer-scale precision under nanomanipulator control. The Si nanowire first served as a nanoprobe to peel a single-layer MoS_2 nanoflake from a triangular nanosheet and transfer it onto another MoS_2 monolayer on a substrate for sliding. Subsequently, the nanowire functioned as a cantilever force sensor to detect friction forces during sliding. The force measured by the cantilever was proportional to its deflection, which could be directly quantified via SEM imaging. Friction tests on sliding between incommensurate MoS_2 monolayers revealed a friction coefficient of

$\approx 10^{-4}$, confirming the regime of superlubricity and highlighting the potential of this integrated approach for studying ultralow-friction interfaces in 2D materials.

3.2.3 STM-based techniques

The interlayer friction testing technology based on scanning tunneling microscopy (STM) shares conceptual similarities with AFM-based nanomanipulation techniques. However, STM offers superior resolution, enabling atomic-scale manipulation capabilities. Its primary limitations include the strict requirement for sample conductivity, susceptibility of probes to wear, and constraints in imaging range compared to alternative methods. These characteristics make STM particularly well-suited for fundamental studies of atomic-scale friction mechanisms, while AFM-based approaches may be more practical for broader material characterization applications.

In 2018, Büch et al.[52] reported the observation of superlubricity in monolayer tungsten disulfide (WS_2) sliding on epitaxial graphene (EG) grown on silicon carbide (SiC). Single-crystalline WS_2 flakes, synthesized via chemical vapor deposition (CVD) with lateral dimensions of hundreds of nanometers, were found to be predominantly aligned with zero azimuthal rotation relative to the EG substrate. When perturbed by a STM tip, the WS_2 flakes exhibited room-temperature sliding on the graphene surface. Atomistic MD simulations based on force fields revealed that localized physical deformation of the WS_2 flake by the tip allowed energy transfer sufficient to overcome the activation barrier for motion, enabling ultralow-friction rototranslational displacement—a hallmark of superlubricity. Post-sliding experiments confirmed that the WS_2 flakes came to rest at orientations rotated by $n\pi/3$ relative to the graphene lattice. High-resolution measurements further demonstrated that the WS_2/EG interface remained atomically sharp, with the WS_2 lattice free from strain after sliding.

4 2D Inorganic Materials in Tribology

Inorganic materials, due to their high-temperature resistance, corrosion resistance, and environmental friendliness, are often used as high-performance lubricants under extreme working conditions, such as the application of ceramic coatings in cutting tools, engine components, and marine equipment. The

crystallization of inorganic materials is relatively easy, leading to the emergence of numerous 2D structures, which play a crucial role in the field of lubrication. For instance, molybdenum disulfide (MoS_2) is often used in aerospace, vacuum machinery, and nuclear industries, while hexagonal boron nitride (h-BN) is commonly applied as the high-temperature insulating lubricants in electronic devices and precision machinery. This section not only presents the latest research progress of traditional 2D lubricating materials with inorganic structure, such as transition metal dichalcogenides (TMDs) and h-BN, but also provides an overview of the tribological research on the emerging 2D inorganic structure, such as 2D transition metal carbides and nitrides (MXenes), Layered Double Hydroxides (LDHs) and Single elements (SEs).

4.1 Transition Metal Dichalcogenides

Transition metal dichalcogenides (TMDs) have the molecular formula MX_2 and possess a sandwich structure of X-M-X, where X represents chalcogen elements (S, Se, or Te), and M indicates transition metal elements from Group IV (Ti, Zr, Hf), Group V (V, Nb, Ta), or Group VI (Mo, W). TMDs can exhibit multiple structural phases. The monolayer of TMDs mainly exist in the trigonal phase (1H), octahedral phase (1T), and distorted octahedral phase (1T'), while bulk TMDs typically have trigonal prismatic phases (2H and 3R), where 1T, 2H, and 3R represent crystal symmetries, namely triangular, hexagonal, and rhombic, respectively[53]. Among these crystal forms, the hexagonal structure is considered to have the lowest friction coefficient. The lattice layers of TMDs are formed by M and X atoms through the strong covalent bonds, while only the weak VDW forces exist between layers. It is widely recognized that the atomic-level thickness and excellent mechanical properties endow TMDs with the low-friction characteristics[54]. The atomic-level thickness of MoS_2 enables low friction through abundant slip planes and prevents abrasive wear, while its exceptional mechanical properties—such as high hardness and compressive strength—ensure durability by withstanding high contact pressures.

The effect of interlayer geometry on the structural superlubricity of TMDs has been a hot topic in both theoretical[55] and experimental[56] research. Li et al.[51] experimentally investigated the tribological properties of monolayer MoS_2 on the MoS_2 substrate using an in-situ scanning electron microscope equipped with a silicon nanowire force sensor, and the superlubricity was achieved (friction coefficient $\mu \approx 10^{-4}$) (Figure 4). In 2018, Liu et al.[45] fabricated a series of probes coated with TMDs that including MoS_2, TaS_2 and ReS_2, and constructed their single-crystalline contacts, so that the interlayer sliding achieved ultra-low friction coefficients. Zhang et al.[57] found that the sliding friction of the AFM probe on $MoTe_2$ surface increased non-monotonically with increasing Te vacancy density in micro-scale experiment, and proved that the non-monotonic variation was determined by the change of the maximum sliding barrier caused by the position change of the tip during the sliding process and the uneven charge distribution caused by charge transfer at defects through the molecular dynamic simulations. The same authors[58] also investigated the surface friction behaviors of $MoTe_2$ with different structures and found that the surface friction force and friction coefficient (μ) of 2H-$MoTe_2$ were one order of magnitude smaller than those of 1T'-$MoTe_2$. The above experimental results were confirmed by Density functional theory (DFT) simulations to be related to the difference in the sliding energy barrier of the potential energy surface (PES) when the tip slid on their surface. Moreover, the tiny energy difference between 2H-$MoTe_2$ and 1T'-$MoTe_2$ provided the possibility of regulating friction by controlling the phase transition. Subsequently, they measured the interlayer friction force by sliding a $MoTe_2$ powder on $MoTe_2$ substrate using an AFM tip[59]. The results showed that the friction coefficient of the 1T'-$MoTe_2$/1T'-$MoTe_2$ interface was 2.025×10^{-4}, lower than that of the 2H-$MoTe_2$/2H-$MoTe_2$ interface (3.086×10^{-4}), while the friction coefficient of the heterogenous interface (1T'-$MoTe_2$/2H-$MoTe_2$) was as low as 6.875×10^{-5}. DFT simulations indicated that the relative magnitudes of the average and the maximum shear strength based on the interlayer potential energy were the reasons for the lower friction force of the 1T'-$MoTe_2$/1T'-$MoTe_2$ interface compared to the 2H-$MoTe_2$/2H-$MoTe_2$ interface. Additionally, the weak interlayer

electrostatic interaction and the reduction of potential energy corrugations due to incommensurability results in the lowest interlayer friction in the heterogenous interface of 1T'-$MoTe_2$/2H-$MoTe_2$. In 2024, Liu's group[47] pioneered the Orienting and Transferring Nanosheets at Phase Interface (OTNPI) technique, a novel nanomanipulation platform that enabled systematic investigation of interlayer friction in molybdenum disulfide (MoS_2), ultimately achieving robust superlubricity states. By synergistically combining molecular friction theory with first-principles DFT calculations, they elucidated the fundamental origins of both interlayer friction and adhesion forces. Furthermore, the team established a computationally efficient energy-based descriptor that successfully correlates with experimentally observed friction coefficient trends.

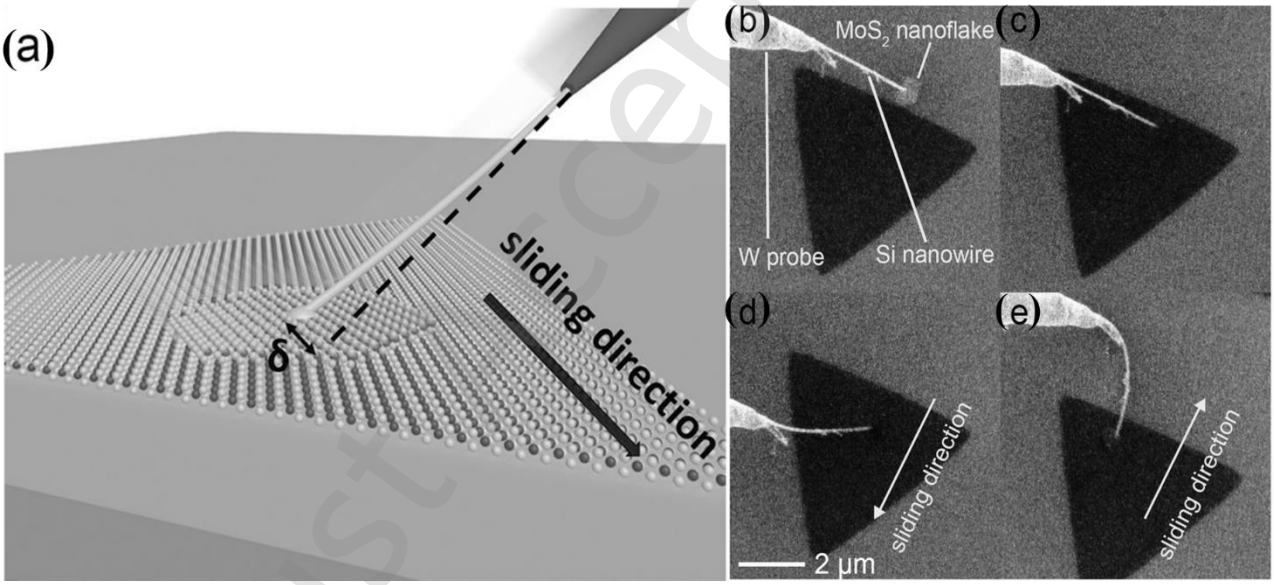

Fig. 4 (a) Schematic diagram of the friction test of a single-layer MoS_2 nanosheet sliding on a single-layer MoS_2 nanosheet substrate. (b-e) SEM images during the friction test process. Reprinted from ref [51] with permission from John Wiley and Sons, Copyright 2017.

In industrial applications, lubricants are essential for reducing friction and wear on contact surfaces like gears, bearings, and reducers. Extensive research has demonstrated that TMDs exhibit outstanding anti-friction and anti-wear performance as lubricant additives, coupled with remarkable thermal stability[60]. The tribological performance of lubricants is predominantly governed by the size, structural characteristics, surface modification, and tribofilm formation behavior of TMDs[61]. The

size-controlled tribological behavior of TMDs additives is well documented by the representative study[62] that the thinner $MoSe_2$ nanosheets derived from the ultrasound process of hydrothermally grown nanoparticles achieve 43% and 87% reductions in friction coefficient and wear rate respectively compared to base oil formulations. Experimental evidence confirms that the formation of protective tribofilms by these nanosheets significantly reduces interfacial friction resistance and wear loss, resulting in markedly improved tribological performance. Nevertheless, the inherent self-aggregation tendency of nanomaterials presents a persistent challenge in obtaining homogeneous dispersion of TMDs nanosheets within lubricating oils, which limits their full lubrication potential. In this regard, surface functionalization emerges as a viable approach to enhance the interfacial compatibility between TMDs and lubricant matrices while attaining optimal dispersion characteristics[63,64]. The incorporation of oleamine (OM)-modified WS_2 nanosheets (WS_2-OM) into polyalphaolefin (PAO6) base oil significantly enhances both dispersion stability and tribological performance, as evidenced by improved anti-friction and anti-wear properties[65]. This dual functionality arises from two distinct mechanisms: (1) the lamellar structure of WS_2-OM enables the formation of a physical protective layer through surface deposition on worn areas, while (2) tribochemical reactions during friction lead to the in-situ generation of a composite protective film consisting of WO_3 and iron oxides. These synergistic effects collectively contribute to the superior lubrication performance compared to the base oil alone. In their 2015 study, Chen et al.[66] demonstrated significant enhancement of extreme pressure properties by incorporating oleylamine-surface-modified MoS_2 nanosheets into liquid paraffin. The addition of merely 1 wt% ultrathin MoS_2 nanosheets resulted in a remarkable improvement in load-bearing capacity - from below 50 N to over 2000 N at 120 °C. Through wear scar analysis, the researchers proposed that this dramatic performance improvement stems from two key factors: the excellent dispersion of the nanosheets and their ultrathin morphology. These characteristics enable the nanosheets to effectively penetrate the contact area between sliding surfaces, where they form a protective film that prevents direct metal-to-metal contact and subsequent seizure. In 2018, Chen et

al.[67] demonstrated that oleylamine-modified ultrathin MoS_2 sheets in base oil could effectively control wear under nominal pressures of approximately 1 GPa. This performance threshold corresponds to the breaking strength of ultrathin MoS_2 when considering the real pressure between surface asperities - a significant improvement over conventional lubricants, which typically fail at just a few hundred megapascals. The researchers conducted comparative studies with MoS_2 sheets of varying lateral dimensions, revealing that larger sheets exhibited superior lubrication performance. The authors attributed these effects to two key factors: (1) the excellent oil solubility and minimal thickness of the nanosheets, facilitating their penetration into asperity contact zones, and (2) the prevention of metal-to-metal contact when real interfacial pressures remain below the material's breaking strength, thereby inhibiting localized seizure and progressive wear. Surface functionalization with oleic acid diethanolamide (OD) induces significant morphological and chemical modifications in 2D-MoS_2 nanosheets (MoS_2-OD) compared to pristine MoS_2[68]. The resulting MoS_2-OD exhibits expanded flake dimensions and forms stable C-S covalent bonds with OD molecules, which dramatically enhances both dispersion homogeneity and colloidal stability in base oil. Tribological evaluations demonstrate remarkable improvements, with the MoS_2-OD composite lubricant achieving 36.8% reduction in friction coefficient and 15.4% decrease in wear rate relative to unmodified base oil. Notably, the friction-reducing performance of MoS_2-OD shows positive correlation with increasing oil viscosity and applied load, suggesting excellent pressure-viscosity adaptability.

Conventional block materials including ceramics, metals, and polymers serve as fundamental components for constructing friction pairs in tribological systems. To enhance their performance[69], TMDs - particularly MoS_2[70], WS_2[71], and $MoSe_2$[72] - have been extensively employed as functional fillers. These TMD nanomaterials are strategically incorporated into various material matrices to optimize tribological characteristics, demonstrating significant improvements in friction reduction and wear resistance[73,74]. Mohammad et al.[75] demonstrated that incorporating 2.5 wt% MoS_2 into a copper matrix significantly improves tribological performance, achieving both reduced

friction coefficient and diminished wear loss. This enhancement primarily stems from the in-situ formation of a protective tribofilm during sliding contact. However, their study revealed a critical concentration threshold, beyond which excessive MoS_2 content (＞2.5 wt%) induces two detrimental effects: (1) progressive softening of the composite material and (2) consequent deterioration of wear resistance properties. In contrast to MoS_2-based composites, Cu/WS_2 systems demonstrate unique tribological behavior under 40% relative humidity conditions. Remarkably, even at high WS_2 loading (40 vol%), the composite maintains its mechanical integrity while achieving substantial friction and wear reduction. This phenomenon originates from shear-induced plastic deformation during sliding, which promotes preferential horizontal alignment of WS_2 nanosheets within the friction layer. The resulting oriented nanostructure effectively combines wear protection with preserved matrix hardness, ultimately yielding superior tribological performance[76]. Despite their excellent tribological properties, 2D-TMDs exhibit significant atmospheric sensitivity, particularly in humid conditions. Elevated humidity levels adversely affect TMD-reinforced composites through two primary mechanisms: (1) enhanced surface adhesion that increases frictional forces, and (2) accelerated formation of detrimental oxide species at sliding interfaces. These humidity-induced phenomena collectively degrade the composite's tribological performance, manifesting as increased friction coefficients and accelerated wear rates. Strategic element doping (e.g., Zr, Al, Au, Ag) and optimized multi-component synergy in composite materials have proven highly effective in counteracting these detrimental effects. These approaches facilitate two critical improvements: (1) mitigation of humidity-induced degradation mechanisms, and (2) enhanced formation of protective tribofilms. Such modifications fundamentally improve the composite's tribological performance by simultaneously reducing friction coefficients (anti-friction) and minimizing material loss (anti-wear), as demonstrated in studies[77-80].

4.2 Hexagonal Boron Nitride

Hexagonal boron nitride (h-BN), commonly referred to as 'white graphite', exhibits a crystalline structure composed of alternating boron and nitrogen atoms arranged in an extended two-dimensional hexagonal lattice. Within each atomic layer, strong sp^2 hybridized bonds with partial ionic character provide exceptional in-plane stability. The interlayer interactions, however, are dominated by weak van der Waals forces combined with relatively large interplanar spacing (~3.3 Å), which facilitates low-energy shear deformation through interlayer sliding. Monolayer hexagonal boron nitride (h-BN) demonstrates remarkable intrinsic properties including exceptional mechanical strength (Young's modulus ≈865 GPa), outstanding thermal conductivity (≈751 $W \cdot m^{-1} \cdot K^{-1}$), and remarkable thermal stability with chemical inertness maintained up to ≈1000°C[81-84]. Intriguingly, multilayer h-BN maintains these superior mechanical characteristics due to strain-induced strengthening of interlayer interactions, which effectively suppresses interlayer sliding[85]. This unique combination of layer-number-independent mechanical robustness, superior thermal properties, and intrinsically low interlayer shear resistance positions h-BN as an ideal candidate for advanced lubrication applications, with demonstrated potential for achieving superlubricity states.

Through first-principles calculations employing dispersion-corrected DFT (DFT-D) with periodic boundary conditions, Kabengele et al.[86] systematically mapped the potential energy surface (PES) governing interlayer sliding in h-BN. Their theoretical investigation revealed a critical phenomenon: once the rotational potential barrier is surmounted, the material exhibits nearly frictionless interlayer sliding within its rotational registry configuration.

Nanoscale h-BN demonstrates superior tribological enhancement compared to its micro- or sub-micrometer counterparts when used as lubricant additives. Experimental evidence reveals that when incorporated into avocado oil, nano-h-BN (≈64% reduction) outperforms micron-sized h-BN (≈8% reduction) in friction coefficient reduction, with corresponding wear rate decreased by 70% versus 13% relative to the base oil[87]. This significant performance enhancement stems from two key mechanisms: (1) nanoscale particles effectively fill surface asperities to form smooth transfer layer,

and (2) interparticle sliding facilitates energy dissipation at the contact interface. Conversely, larger particles may induce detrimental third-body abrasion through interfacial plastic deformation. The poor dispersibility of h-BN in lubricants necessitates surface modification for its practical application as an additive. Fluorination treatment emerges as an effective approach, with fluorinated boron nitride nanosheets (F-BNNSs) demonstrating remarkable tribological enhancement[88]. These modified nanosheets exhibit superior dispersion stability compared to pristine h-BN and undergo morphological transformation to nanorod structures at elevated fluorine concentrations. Optimal F-BNNSs loading achieves substantial friction reduction (32.3%) and exceptional wear resistance improvement (>91%). The lubrication mechanism involves: (1) rapid friction reduction through the shear-alignment of nanosheets and nanorod-nanosheet hybrid structures, and (2) wear protection via formation of a composite tribofilm comprising chemically reacted layers and physically deposited materials, working synergistically with the self-lubricating nanostructures. A breakthrough in macroscopic superlubricity was achieved by incorporating ionic liquids (ILs) and hydroxylated hexagonal boron nitride nanosheets (HO-BNNs) into ethylene glycol-water solution (EGaq), demonstrating persistent and robust superlubricating behavior at steel-steel point contacts[89]. This remarkable performance originates from the synergistic interaction between three key components: (1) EG molecules and ILs form an adsorbed boundary film through tribochemical reactions; (2) hydrogen-bonded HO-BNNs maintain stable dispersion while continuously supplying the contact interface; and (3) the combined action generates multiple beneficial effects including mechanochemical reactions, in-situ surface polishing, and self-cleaning. The resulting composite tribofilm serves dual functions: physically separating asperity contacts to prevent direct surface interaction, while chemically passivating the interface to inhibit pit propagation and wear progression.

Uncoated substrates are particularly vulnerable to tribological damage, exhibiting pronounced adhesion, abrasive scratching, and oxidative degradation during frictional contact[90]. Recent investigations have demonstrated the remarkable efficacy of h-BN as a reinforcing filler in aromatic

thermosetting polymer (ATSP) coatings for steel substrates[91]. The incorporation of 10 wt% h-BN into ATSP coatings yields dramatic improvements - reducing the friction coefficient by 86% and wear volume by 70% compared to bare substrates. This exceptional performance arises from four synergistic mechanisms: (1) Thermal management - h-BN's high thermal conductivity dissipates frictional heat, mitigating thermal degradation and oxidation; (2) Mechanical reinforcement - surface pretreatment combined with h-BN creates a composite architecture with enhanced load-bearing capacity[92]; (3) Tribological modification - h-BN's lamellar structure facilitates interlayer shear, minimizing adhesive wear and abrasive particle generation; (4) Surface engineering - h-BN participates in tribochemical reactions to form a protective boundary film that sustains lubrication under severe conditions. While h-BN demonstrates excellent friction-reducing and anti-wear properties in composite coatings, its practical application faces significant challenges due to the intrinsic characteristics of h-BN nanosheets. The high surface energy and absence of active functional groups on h-BN nanosheets lead to severe particle agglomeration and poor interfacial bonding with the matrix material[93,94]. These agglomerates create structural defects and localized stress concentrations within the coating. Under cyclic frictional loading, the weakly bonded h-BN clusters are preferentially detached from the coating surface, ultimately compromising the tribological performance[92]. This particle pull-out phenomenon not only reduces the coating's durability but also generates third-body abrasives that accelerate wear degradation. Li et al.[95] developed an effective surface modification strategy to improve the dispersion ability of h-BN in epoxy (EP) matrices through NaOH-mediated hydroxylation (OH-h-BN). The resulting EP/OH-h-BN composites exhibited remarkable mechanical enhancement, showing increased storage modulus and tensile strength relative to pure EP. Tribological testing revealed significant performance improvements, with the composite coating demonstrating 27% lower friction coefficient and 58% reduced wear rate compared to unmodified EP. These enhancements stem from three key mechanisms: (1) The hydroxylated surface enables uniform OH-h-BN dispersion, facilitating effective transfer to the friction interface where it forms a continuous tribofilm; (2) Friction-induced

heating promotes the formation of beneficial tribochemical products (B_2O_3, FeO, Fe_2O_3) that reinforce the film's mechanical properties[96,97]; (3) Strong interfacial bonding between OH-h-BN and the EP matrix inhibits microcrack initiation and propagation (Figure 5). The synergistic combination of these effects creates an effective barrier that prevents direct steel-coating contact while maintaining coating integrity under shear stress.

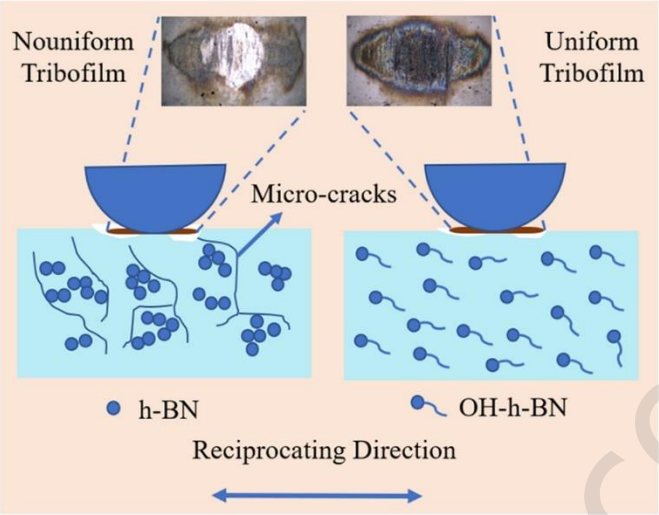

Fig. 5 The friction reduction mechanisms of h-BN and OH-h-BN. Reprinted from ref [95] with permission from Elsevier, Copyright 2022.

4.3 Metal Carbene/Nitrene Compounds

A new class of 2D inorganic materials called MXenes (layered transition metal carbides and nitrides) has recently gained significant attention in tribology research[28]. These materials follow the general formula $M_{n+1}X_nT_x$ (where n = 1-4), in which M denotes transition metals (e.g., Ti, Zr, Nb, Mo, Mn), X represents carbon and/or nitrogen atoms, and T_x signifies surface functional groups such as -O, -OH, -F, or -Cl[98]. MXenes are typically synthesized through selective etching of their parent MAX phases, represented by the general formula $M_{n+1}AX_n$ (n = 1-4). In this structure, M and X maintain the same definitions as in MXenes (transition metals and carbon/nitrogen respectively), while A denotes an A-group element (e.g., Al, Si, or Zn)[99]. This etching process enables MXenes to partially retain the outstanding mechanical strength and thermal stability characteristic of their MAX phase precursors[100]. Extensive research has confirmed that the surface terminations and atomic

configurations of MXenes play a crucial role in modulating their mechanical and tribological properties[101].

Recent studies have systematically explored the interfacial properties of Ti-based MXenes. Righi et al.[102] theoretically demonstrated that surface terminations play a critical role in determining the interlayer and substrate adhesion characteristics of these materials. Subsequent experimental investigations by Rodriguez et al.[101] utilized friction force microscopy (FFM) to characterize $Ti_3C_2T_x$ nanosheets deposited on silica substrates, producing detailed friction maps that highlighted the exceptional potential of few-layer $Ti_3C_2T_x$ as a nanoscale solid lubricant. Building upon this foundation, Mochalin et al.[103] conducted comprehensive comparative analyses in 2022, examining the nanoscale frictional behavior of various MXenes ($Ti_3C_2T_x$ and Ti_2CT_x) and in contact with different counter-surfaces such as graphene and $MoSe_2$. Their findings revealed that specific surface terminations (-O and -OH groups) induce hydrogen bonding at the interface, thereby significantly enhancing frictional forces during MXenes/MXenes interactions. Recent experimental studies[104] have demonstrated the remarkable tribological performance of Ti_3C_2 MXenes coatings. When deposited on SiO_2 substrates and tested against diamond-like carbon (DLC) coated steel balls under dry N_2 conditions, these coatings exhibited exceptional superlubricating properties, achieving an ultra-low friction coefficient of 0.0017 - approximately 3.3-fold reduction compared to bare silicon substrates. This outstanding performance primarily stems from two key factors: the chemically inert nature of the DLC counterface and the unique graphene-like interlayer shear mechanism of the Ti_3C_2 MXenes. However, the study also revealed a critical limitation: the unpassivated MXenes surface tends to undergo tribochemical reactions during prolonged sliding, which progressively degrades the lubricating performance and eventually leads to significant friction increase and accelerated wear of the DLC coating.

The effectiveness of MXenes as lubricant additives critically depends on their uniform dispersion within the base oil to achieve optimal anti-friction and anti-wear performance[105]. However, the

inherent surface chemistry of MXenes nanosheets - characterized by abundant functional groups (-OH, -O, and -F) - combined with their exceptionally high specific surface area, leads to elevated surface energy and pronounced hydrophilicity. These intrinsic properties significantly compromise the dispersion stability of MXenes in hydrophobic lubricating oils, as evidenced by previous studies[106,107]. Consequently, developing effective strategies to enhance MXenes dispersion represents a crucial research direction for maximizing the tribological benefits of MXenes-based lubrication systems. Extensive research has been conducted to evaluate the concentration-dependent tribological behavior of MXenes in lubrication systems and their load-responsive friction characteristics[105]. Systematic investigations reveal that Ti_3C_2 MXenes additives significantly enhance both friction reduction and wear resistance compared to pure paraffin oil. The optimal performance is achieved at 1.0 wt% concentration, demonstrating superior tribological properties. However, when the concentration exceeds approximately 6.0 wt%, the friction coefficient markedly increases due to the agglomeration of 2D Ti_3C_2 nanosheets. Two distinct lubrication mechanisms have been identified: (1) the formation of a protective oil film dominates at lower loads (8-20 N), while (2) the development of a friction-induced tribofilm becomes predominant under higher loads (20-30 N). Surface functionalization has emerged as an effective strategy to enhance the compatibility of MXenes with base lubricants[24,108-110]. A representative example[107] involves the modification of Ti_3C_2 nanosheets with tetradecylphosphonic acid (TDPA), yielding TDPA-functionalized Ti_3C_2 (TDPA-Ti_3C_2) that demonstrates excellent dispersion stability in castor oil. Tribological evaluations reveal remarkable performance improvements: the 0.1 wt% TDPA-Ti_3C_2/castor oil system exhibits 27.9% reduction in friction coefficient and 55.1% decrease in wear rate compared to pure castor oil. More significantly, when benchmarked against unmodified 0.1 wt% Ti_3C_2/castor oil, the TDPA-modified counterpart shows further enhancements of 22.5% in friction reduction and 62.4% in wear resistance. This substantial performance improvement can be attributed to three synergistic mechanisms: (1) the long alkyl chains of TDPA promote molecular-level compatibility with the oil matrix through

hydrophobic interactions; (2) intercalation of phosphate groups expands the interlayer spacing of Ti_3C_2, facilitating exfoliation and dispersion; and (3) the well-dispersed nanosheets readily access the friction interface, where they form a robust protective tribofilm that prevents direct contact between sliding surfaces while effectively supporting applied loads. The surface functional groups of MXenes significantly improve their compatibility with water/alcohol-based lubricating systems. When a $Ti_3C_2T_x$/glycerol mixture is introduced at the Si_3N_4/sapphire interface, it enables the achievement of macroscopic liquid superlubricity, demonstrating an exceptionally low friction coefficient of approximately 0.002 under demanding conditions of 79.1 MPa contact pressure and 15% relative humidity[111]. This remarkable performance results from a synergistic combination of two lubrication mechanisms operating in parallel: (1) In boundary lubrication regime, tribochemical reactions between Si_3N_4 and $Ti_3C_2T_x$ nanosheets during the running-in period lead to the formation of a protective tribofilm on the Si_3N_4 surface. The inherent low interlayer shear strength of MXenes facilitates a sliding mechanism transition from the original ceramic interface to the weakly bonded MXenes interlayers. (2) In elastohydrodynamic lubrication regime, the abundant hydroxyl groups in both glycerol and water molecules form strong hydrogen bonds with $Ti_3C_2T_x$ nanosheets, creating a well-hydrated surface layer that substantially reduces the shear strength of the lubricating fluid film. The same lubrication mechanism has been successfully demonstrated in a Mo_2CT_x-enhanced lithium hexafluorophosphate ionic liquid lubrication system at the Si_3N_4/sapphire interface[112]. Remarkably, this system achieves an unprecedented breakthrough by extending the critical contact pressure for maintaining macroscopic superlubricity to approximately 1420 MPa - representing a significant advancement in extreme-pressure lubrication performance.

MXenes have recently gained significant research interest as solid lubricant coatings and reinforcement components in composite coatings[113,114]. Theoretical studies using DFT demonstrate that MXenes exhibit excellent dispersion within coating matrices while simultaneously improving interfacial bonding stability with substrates - properties that collectively enhance the overall

protective performance of these coatings[115]. Recent studies[116] have systematically examined the effects of contact pressure and relative humidity on the tribological performance of $Ti_3C_2T_x$ coatings. Under optimized conditions (0.8 GPa contact pressure and 20% relative humidity), the MXenes coating demonstrated remarkable improvements over the bare substrate, exhibiting a 2.3-fold reduction in friction coefficient and a 2.7-fold decrease in wear rate. These significant enhancements can be attributed to the in-situ formation of a dense, protective tribofilm consisting of well-aligned MXenes nanosheets during the friction process. Follow-up investigations[117] demonstrated the tribological performance of drop-cast MXenes coatings in thrust ball bearing applications under specific environmental conditions (0.8 GPa contact pressure, 50% relative humidity). The coated specimens exhibited superior performance metrics compared to their uncoated counterparts, showing a 3.2-fold reduction in friction torque, a 2.1-fold increase in service life, and a 2.9-fold decrease in linear cumulative wear rate. Tribological evaluation of 100 nm-thick electrosprayed multilayer $Ti_3C_2T_x$ coatings was performed under dry conditions at 0.3 GPa contact pressure[118]. The optimized MXenes coating demonstrated exceptional performance, exhibiting a sixfold reduction in friction coefficient and achieving an ultralow wear rate of 4×10^{-9} $mm^3/N\cdot m$ after 100,000 sliding cycles (Figure 6a). This remarkable tribological enhancement stems from the formation of a protective tribofilm comprising thermally/mechanically degraded MXenes nanosheets and amorphous/nanocrystalline iron oxides, which effectively prevents direct surface-to-surface contact during sliding (Figure 6b). The formation of a stable tribofilm represents a critical mechanism for enhancing tribological performance in MXenes-based coatings. The inherent layered structure of MXenes offers substantial surface area, while the abundant titanium atoms facilitate surface functionalization through bonding with -OH and -F groups, thereby strengthening the coating-substrate interface. Experimental studies demonstrate this principle through MXenes-EG composite coatings, where uniform dispersion of MXenes nanosheets in ethylene glycol (EG) followed by deposition on steel substrates yields superior performance[119]. Under testing conditions of 6 N load and 5 Hz sliding frequency, the composite coating achieves a

significantly reduced friction coefficient (μ≈0.165) compared to pure MXenes coatings, while demonstrating exceptional long-term stability. The EG additive plays a multifunctional role by: (1) improving MXenes dispersion stability, (2) enhancing interfacial shear strength, and (3) strengthening coating-substrate adhesion. These synergistic effects promote the formation and retention of an effective transfer film, which minimizes direct surface contact and enables the simultaneous achievement of ultralow friction and exceptional wear resistance. In a recent breakthrough, Yang et al.[120] successfully fabricated Ti-MXene hybrid nanomembranes consisting of in-situ chemically bonded metallic Ti and $Ti_3C_2T_x$ MXene nanocrystals. These nanomembranes demonstrated exceptional isotropic mesoscale superlubricity, exhibiting an ultra-low coefficient of friction (<0.01) and near-zero wear (wear rate $<10^{-9}$ mm^3/Nm) under extreme conditions, including contact pressures exceeding 10 GPa and elevated temperatures ranging from 473 to 573 K. This remarkable performance stems from the confinement of MXene within Ti nanocrystals, which simultaneously enables self-lubrication and enhances wear resistance by reducing the energy barrier for tribochemical protection formation.

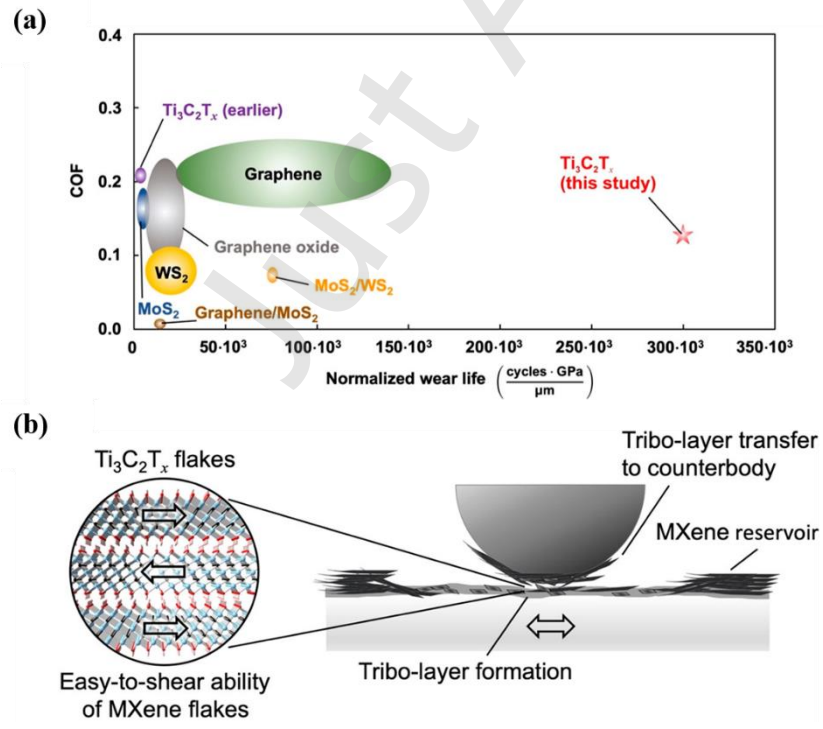

Fig. 6 (a) Comparative evaluation of normalized wear life and friction coefficients for MXenes versus other 2D materials serving as solid lubricants under identical testing conditions. (b) Proposed mechanistic explanation for the simultaneous achievement of exceptional wear resistance and sustained low friction in MXene-based lubricants. Reprinted from ref [118] with permission from ACS publications, Copyright 2021.

4.4 Layered Double Hydroxides

Layered double hydroxides (LDHs) feature a unique architecture consisting of positively charged metal hydroxide layers interspersed with interlayer anions (e.g., CO_3^{2-}, NO_3^-) and water molecules. Their tunable chemical composition, adjustable interlayer spacing, and customizable anion types/quantities confer exceptional versatility for lubrication applications[121]. For example, functional anions (e.g., corrosion inhibitors or antioxidants) can be incorporated via intercalation modification to enhance lubricating grease performance. A distinctive "memory effect" further distinguishes LDHs: calcination at 450~500 °C transforms them into composite metal oxides, which subsequently regenerate their layered structure upon exposure to specific anion solutions. This property offers potential self-repair capabilities in high-temperature lubrication systems. The LDH structure contains abundant hydroxide ions (OH^-), intercalated carbonate ions (CO_3^{2-}), and crystalline water. Upon thermal exposure, these components decompose to release H_2O and CO_2, which collectively contribute to flame retardation through three mechanisms: (1) dilution of combustible gas concentrations, (2) formation of an oxygen-deprivation barrier, and (3) endothermic cooling. This synergistic action markedly improves the fire-resistant properties of lubricating greases. Additionally, LDHs' amphoteric nature (acidic/basic dual sites) neutralizes acidic oxidation byproducts, mitigating oil degradation and prolonging grease service life.

Through combined DFT calculations and ab initio molecular dynamics (AIMD) simulations, Tran et al.[122] systematically investigated the friction-reduction mechanisms in Mg_2Al_1-LDH systems. Their computational studies revealed two key findings: (1) The incorporation of trivalent cations

significantly enhances the lubricating performance of LDH materials, and (2) The lateral force exhibits a pronounced dependence on both surface hydroxyl group coverage and interfacial water/anion layer thickness, with thicker interfacial layers leading to reduced friction. Wang et al.[123] systematically investigated the nanotribological properties of LDH nanosheets with three distinct surface microstructures, namely LDH-Mg, LDH-Zn and LDH-Co, all of which exhibited exceptional superlubricity characteristics. Their study revealed a significant coordination-dependent friction behavior: when the divalent metal ions transitioned from tetrahedral to octahedral coordination, the system demonstrated a two-fold increase in friction coefficient (from 1.0×10^{-3} to 3.9×10^{-3}) accompanied by a 62.3% enhancement in adhesion force. Through combined crystal field theory (CFT) analysis and DFT calculations, the researchers identified two fundamental mechanisms underlying the superlubricating performance: (1) pronounced interfacial charge density transfer between the LDH nanosheets and AFM probe, and (2) formation of an ultra-low sliding energy barrier interface between LDHs and highly oriented pyrolytic graphite (HOPG) substrates.

In their 2017 study, Wang et al.[124] systematically investigated the tribological performance of NiAl-LDH nanoplatelets as lubricant additives in base oil using a ball-on-disk reciprocating tribometer. The experimental results revealed two significant findings under extreme contact pressures (up to 2.16 GPa): (1) the addition of nano-LDHs led to a 10% reduction in the coefficient of friction (COF) coupled with substantial wear resistance enhancement, and (2) larger-sized nanoplatelets exhibited superior and more stable tribological performance compared to their smaller counterparts. Comprehensive surface characterization and structural analysis indicated that the improved performance was attributed to the higher crystallinity of the nanoplatelets, which facilitated the in-situ formation of a mechanically robust tribofilm during the sliding process. In 2020, Wang et al.[125] developed a series of LDH nanoplatelets with precisely controlled thickness variations. The researchers particularly focused on ultrathin Co/Al-LDH (U-CA-LDH) nanoplatelets (≈1 nm thickness, corresponding to 1-2 atomic layers), which were surface-functionalized with ethylene glycol to ensure

stable colloidal dispersion in oil-based lubricants. Remarkably, the U-CA-LDH modified lubricant demonstrated exceptional extreme pressure performance, sustaining loads up to 2000 N - approximately threefold higher than conventional commercial additives. Detailed tribofilm characterization revealed that U-CA-LDH nanoplatelets with partial tetrahedral coordination exhibited superior tribological properties compared to multilayer Mg/Al-LDH (M-MA-LDH) with perfect octahedral coordination. The authors attributed this enhancement to the defective coordination structure of U-CA-LDH, which (1) provided highly active sites for interfacial reactions under tribological stress, and (2) facilitated the generation of a compact, mechanically robust protective film on sliding surfaces through thermally activated surface reconstruction processes. In their concurrent 2020 study, Wang et al.[126] pioneered an innovative approach integrating catalytic and tribological functionalities through calcined LDH derivatives. Through controlled thermal treatment of NiAl-LDH under distinct atmospheres, they successfully synthesized nickel oxide (via air calcination, denoted as LDH-C-Air) and metallic nickel nanoparticles (via argon calcination, LDH-C-Ar). Remarkably, the lubricant formulation containing merely 1 wt% LDH-C-Air additives demonstrated exceptional wear resistance, showing negligible surface degradation after prolonged (2 h) sliding tests under extreme contact pressure (~637 MPa). This outstanding performance was attributed to the in-situ formation of a mechanically enhanced tribofilm (~60 nm thickness) that effectively shielded the substrate from wear damage. In their 2021 study, Wang et al.[127] systematically engineered LDHs with tailored chemical compositions (incorporating Co^{2+}, Mg^{2+}, Zn^{2+}, and Ni^{2+} cations) and precisely controlled morphologies (including flower-like, spherical, and plate-like architectures). These nanostructured additives were homogeneously dispersed in base grease and subjected to rigorous tribological evaluation using a ball-on-disk tribometer under ultrahigh contact pressure (2.47 GPa). The flower-like LDH variant exhibited exceptional performance metrics, reducing wear volume to merely 0.2% of the baseline grease value at an optimal concentration of 1 wt%. Advanced characterization techniques revealed that this superior performance originated from the formation of a unique tribofilm featuring: (1) hierarchical

microstructural organization, (2) homogeneous elemental distribution, and (3) enhanced mechanical properties - all resulting from controlled tribochemical reactions between the LDH additives and sliding interfaces. The surface of LDHs features abundant hydroxyl groups, which confer a natural affinity for water and alcohol. In 2019, Wang et al.[128] synthesized two distinct forms of LDHs and dispersed them in polyalkylene glycol (PAG) aqueous solutions: ultrathin nanosheets (ULDH-NS) with dimensions of approximately 60 nm in width and 1 nm in thickness (single or double layer), and nanoparticles (LDH-NP) measuring around 19.73 nm in width and 8.68 nm in thickness. The results demonstrated that the addition of ULDH-NS shortens the running-in period required to attain superlubricity by as much as 85% (achieved within 1000 s) while simultaneously increasing the ultimate load-bearing capacity by a factor of four (reaching 92.7 MPa). Due to their ultrathin longitudinal dimensions, ULDH-NS do not disrupt the fluid film coverage in the contact zone compared to the lubricating PAG solution. Furthermore, AFM analyses demonstrated that ULDH-NS adsorption polishes and protects solid surfaces, attributed to their weak interlayer interactions and enhanced adhesion effects. In 2023, Wang et al.[129] successfully demonstrated ultrafast macroscale superlubricity ($\mu \approx 0.006$) using a novel lubricating system comprising layered double hydroxides (LDHs) dispersed in an ionic liquid-alcohol solution (1-butyl-2,3-dimethylimidazolium tetrafluoroborate [BMMIm][BF_4]). This achievement was realized at the engineering interface between polytetrafluoroethylene (PTFE) and bearing steel, with an exceptionally short running-in period (<7 s). Comprehensive experimental characterization and theoretical analysis revealed that this remarkable superlubricity performance stems from: (1) tribochemical reactions facilitating the formation of nanostructured adsorption layers, where LDHs strongly adhere to the bearing steel surface, and (2) subsequent transformation of the sliding interface into a PTFE-LDH heterogeneous system (Figure 7).

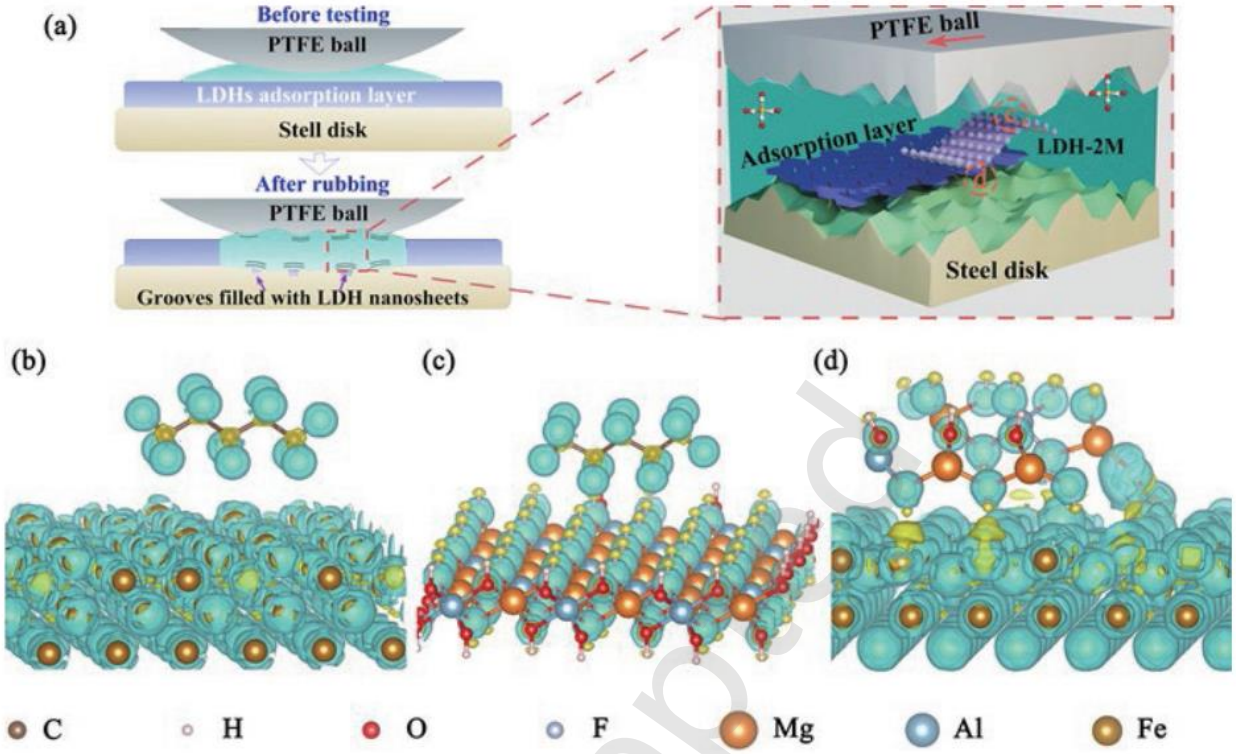

Fig. 7 (a) Proposed mechanism for liquid superlubricity mediated by LDH nanosheets at the PTFE–bearing steel interface. DFT-calculated difference charge density at the (b) PTFE–Fe, (c) PTFE–LDH, and (d) LDH–Fe interfaces providing theoretical evidence. Reprinted from ref [129] with permission from Tsinghua University Press, Copyright 2023.

In their pioneering 2013 study, Guo et al.[130] successfully developed exfoliated polymer/LDH nanocomposites through a sophisticated modification process. The researchers first functionalized nanoplatelet-like LDH with N-Lauroyl-glutamate (denoted as LDH-LG), which was subsequently blended with polyester acrylate resin and cured via UV irradiation. This innovative approach yielded nanocomposite films with dramatically enhanced wear resistance. Remarkably, at just 2 wt% loading of LDH-LG, the nanocomposite exhibited substantial improvements in tribological properties: an 87% reduction in static friction coefficient, a 92% decrease in dynamic friction coefficient, and a 65% reduction in scratch width compared to the unmodified polyester acrylate matrix. In their 2023 study, Diao et al.[131] incorporated 2D calcium aluminum layered double hydroxides (Ca/Al-LDHs) as solid lubricant additives to significantly improve the wear resistance of PTFE-based composites.

Experimental data revealed that the LDH-PTFE composite with an optimal Ca/Al-LDHs loading of 8 wt% exhibited a 3.4-fold enhancement in wear resistance compared to pure PTFE, while preserving its intrinsic mechanical properties. This remarkable improvement was mechanistically attributed to: (1) the inherent lubricity of Ca/Al-LDHs, and (2) hydrogen-bond interactions between OH^- anions in Ca/Al-LDHs and -F group in PTFE, which collectively facilitated the formation of a Ca/Al-LDHs-rich protective transfer film during frictional processes. In a recent study, Guo et al.[132] developed a novel hierarchical nanocomposite through a multi-step modification process. Initially, zirconium phosphate (ZrP)-functionalized Zn-Al layered double hydroxides (ZrP-LDHs, denoted as ZL) were synthesized. These ZL nanoparticles were subsequently modified with tannic acid (TA) and cerium(III) ions to obtain ZrP-LDH-PTA-Ce(III) (TCZL) nanohybrids. When incorporated into waterborne epoxy (WEP) coatings, these TCZL nanofillers significantly enhanced the tribological performance. The optimized TCZL/WEP composite coating demonstrated remarkable dual functionality, combining exceptional wear resistance (showing a 75.27% reduction in wear rate compared to pure WEP) with superior corrosion protection. Electrochemical impedance spectroscopy (EIS) analysis revealed that the $|Z|_{0.01Hz}$ value of TCZL/WEP coating maintained a two-order-of-magnitude advantage over the unmodified WEP coating after 35 days of immersion testing, confirming its outstanding long-term anti-corrosion performance.

4.5 Single elements

2D Single-element (2D-SE) materials, defined as atomically thin crystals composed exclusively of one elemental species[12,133,134], represent a unique class of allotropes distinguished from their bulk counterparts. Within the field of tribology, four archetypal systems have demonstrated exceptional performance: graphene[135], silicene[136], phosphorene (black phosphorus[137] and violet phosphorus[138] allotrope), and antimonene[139]. Given graphene's paradigmatic status in tribological research – evidenced by its unparalleled mechanical properties and extensive application records – it is conventionally categorized under carbon-based nanostructures and will be systematically

addressed in subsequent sections. The present discussion therefore concentrates on non-carbon 2D-SE materials, all of which exhibit characteristic inorganic structural frameworks. These 2D-SE materials exhibit a characteristic hexagonal honeycomb lattice configuration, which facilitates interlayer shear deformation owing to their exceptionally low exfoliation energy (typically < 50 meV/atom). This unique combination of structural and energetic properties renders them particularly suitable for diverse tribological applications requiring ultralow friction coefficients.

The crystallographic planes of non-carbon 2D-SE materials frequently display surface roughness at the atomic scale, in contrast to the pristine flatness characteristic of graphene. This morphological distinction arises from variations in valence electron configurations among different elements, which fundamentally alter their bonding geometries and surface energetics. For instance, the structural configuration of black phosphorus (BP) features each phosphorus atom forming three covalent bonds with adjacent atoms within the atomic plane, creating a distinctive puckered honeycomb lattice. This unique folded geometry (with a characteristic interlayer distance of ~5.3 Å) confers exceptional mechanical resilience, making BP particularly advantageous for tribological applications under extreme operational conditions, including high-load scenarios where conventional lubricants typically fail[140,141]. The structural anisotropy inherent in this configuration allows for directional stress dissipation, significantly enhancing its performance in boundary lubrication regimes. In 2016, Chen et al.[142] performed a quantitative characterization of the mechanical properties of suspended black phosphorus (BP) nanoribbons using AFM-based nanoindentation techniques. Their systematic study revealed a pronounced dependence of the elastic moduli on the relative orientation between the nanoribbon's crystallographic lattice and the supporting groove. Theoretical modeling based on classical elasticity theory yielded ideal elastic modulus values of approximately 65 GPa and 27 GPa for zigzag- and armchair-oriented BP nanoribbons, respectively. These experimental findings significantly advanced the fundamental understanding of BP's intrinsic anisotropic mechanical behavior at the nanoscale. Li et al.[141] systematically investigated the atomic-scale frictional

behavior and underlying mechanisms of BP under varying compressive loads using first-principles calculations. Their study revealed that BP exhibits remarkable load-insensitive friction characteristics compared to graphene, a phenomenon attributed to its intrinsic negative Poisson's ratio (-0.027 along the armchair direction) as confirmed by tribological simulations. Furthermore, the interlayer and intralayer electron density redistribution in BP facilitates efficient energy dissipation through charge transfer mechanisms during high-load sliding, thereby achieving superior friction mitigation. These fundamental insights provide crucial guidance for developing next-generation solid lubricants incorporating 2D-SE nanomaterials, particularly for extreme-pressure applications. In 2017, Cui et al.[143] conducted a comprehensive study on the thickness-dependent and anisotropic atomic-scale frictional characteristics of BP. Their research revealed that BP flakes exceedingly approximately five layers exhibited frictional behavior comparable to bulk material, while thinner samples demonstrated a progressive increase in friction with decreasing layer number - a phenomenon attributed to the strengthening mechanism. The study identified pronounced frictional anisotropy, with the armchair orientation displaying maximum friction, the zigzag direction showing minimum values, and intermediate orientations exhibiting transitional behavior. These observations were rationalized through analysis of the anisotropic tip-induced flexural deformation amplitudes in BP flakes. Gong et al.[144] conducted MD simulations to systematically investigate the nanoscale frictional anisotropy of phosphorene, employing a diamond tip sliding across a silicon-supported phosphorene surface along various crystallographic direction. Their simulations revealed a distinctive "M-shaped" frictional profile, characterized by local maxima at 15° and 75° orientations relative to the zigzag direction. This anisotropic behavior was attributed to orientation-dependent potential energy variations stemming from phosphorene's unique puckered structure and its heterogeneous interfacial interactions with the underlying substrate. Wu et al.[145] systematically characterized the enhanced lubrication properties of ambient-degraded BP through AFM measurements coupled with energy-dispersive X-ray spectroscopy (EDS) mapping. Their experimental results demonstrated a remarkable ~50% reduction

in frictional force within the degraded regions of BP nanosheets. The researchers proposed that this friction reduction mechanism primarily stems from the synergistic effects of adsorbed water molecules and the formation of P-OH functional groups on the oxidized BP surface, which collectively modify the interfacial interaction properties. Similarly, a superlubricating interface between the degraded BP flake and the SiO_2/Si substrate was observed[146]. Quantitative measurements demonstrated an exceptionally low interfacial shear stress (ISS) of 0.029 ± 0.004 MPa, which approaches the theoretical limit observed in incommensurate rigid crystalline systems. Structural characterization of the interface identified that the presence of confined water layers between the degraded BP and substrate surface played a crucial role in achieving this ultralow friction state. The molecularly thin water film effectively reduced shear resistance through its unique rheological properties at the nanoscale interface. In their 2024 study, Zhang et al.[147] achieved microscale superlubricity ($\mu \approx 0.0086$) by sliding a diamond-like carbon (DLC) AFM microsphere probe against a relatively rougher black phosphorus (BP) composite surface. This phenomenon was attributed to BP degradation under oxygen and moisture exposure, generating phosphorus oxides (oBP) with abundant P=O and P-OH functional groups. Comprehensive characterization revealed that these oxides, along with a locally graphitized amorphous carbon film at the sliding interface, effectively minimized direct asperity contact while providing ultralow shear resistance—key factors enabling superlubricity. Moreover, the engineered nanoscale surface roughness of the composite enhanced lubrication stability by (1) increasing the effective contact area and (2) generating capillary forces that anchored the water-absorbed oBP lubricating film.

In 2021, Tang et al.[148] demonstrated that 2D black phosphorus (BP) nanoparticles, serving as oil-based nanoadditives in oleic acid (OA), enable macroscale superlubricity for steel/steel contacts under high pressure (with a maximum Hertz contact pressure of 1.1 GPa). The tribosystem lubricated by OA containing 0.1 wt% BP nano-additives (nano-BP-OA) exhibited significantly enhanced tribological performance. Compared to pure OA (COF ≈ 0.09), the nano-BP-OA achieved a substantially lower

coefficient of friction (COF) of 0.006. A remarkable reduction in wear rate was also observed: the wear rate for nano-BP-OA was 1.32×10^{-7} mm^3/N·m, while that for pure OA was 7.78×10^{-6} mm^3/N·m. This outstanding performance is attributed to tribochemical mechanisms whereby BP rapidly captures carboxylic groups from OA. Under the combined effects of high contact pressure and frictional heating, OA decomposes to release passivating species. These species recombine with BP to form an amorphous carbon-rich composite solid tribofilm. Concurrently, OA and passivating groups adsorb onto this tribofilm, creating a passivating layer that ultimately results in macroscale superlubricity. These mechanisms were elucidated through characterization of the tribofilm and Reactive Molecular Dynamics (RMD) simulations. These findings are further corroborated by Gao et al.'s subsequent investigation[149], which demonstrated macroscale superlubricity in engineering materials through the incorporation of partially oxidized black phosphorus (oBP) in oleic acid (OA) lubricating media. Their comparative analysis revealed that the phosphorus oxides generated via active oxidation process exhibited superior tribological performance to pristine BP particles, manifesting in both significantly reduced friction coefficients and enhanced deposition kinetics at sliding interfaces. Building upon these findings, Gao et al.[150] developed an innovative approach for achieving self-adaptive, oil-based macroscale superlubricity across diverse tribopairs, notably steel-steel and steel-diamond-like carbon (DLC) interfaces. This breakthrough methodology leverages two synergistic mechanisms: (1) the exceptional lubrication performance of actively oxidized black phosphorus (oBP), and (2) the surface-mediated catalytic cleavage of oil molecules on oBP nanostructures, which collectively enable dynamic friction modulation under varying operational conditions. Surface-modified BP demonstrates promising potential as an effective additive for water-based lubricants. In their 2018 study, Wang et al.[151] successfully demonstrated robust superlubricity in aqueous environments using NaOH-modified BP (BP-OH) nanosheets. The system exhibited exceptional performance across broad operational parameters, including varying additive concentrations (1-7 wt%), contact pressures (0.5-1.0 GPa), and sliding velocities (56-282 mm/s), achieving an ultralow coefficient of friction (COF) of

0.0006. This remarkable lubrication behavior was primarily attributed to the formation of a highly fluid water boundary layer between the BP-OH nanosheets, which significantly reduced interfacial shear resistance (Figure 8). In 2020, researchers achieved stable dispersion of black phosphorus quantum dots (BPQDs) in ethylene glycol (EG), forming a homogeneous BPQDs-EG aqueous suspension (BPQDs-EG(aq))[152]. Remarkably, this nanofluid enabled macroscale superlubricity (μ = 0.002) at the Si_3N_4/sapphire tribological interface under extreme contact pressure (336 MPa). Comparative wear analysis revealed that the BPQDs-EG(aq) system reduced the wear rate to merely 5.96% of that observed with pure EG aqueous solution (EG(aq)), highlighting the significant anti-wear enhancement. This performance improvement stems from two synergistic mechanisms: (1) the nanoscale rolling effect of BPQDs and (2) the ultralow interlamination shear stress within BPQD stacks. Furthermore, the in-situ formation of phosphorus oxides (P_xO_y) during friction contributed to the stable maintenance of superlubricity by passivating surface asperities and facilitating shear localization. In their groundbreaking 2021 study, Ren et al.[153] demonstrated exceptional liquid superlubricity ($\mu \approx 0.004$) under extreme contact pressures up to 1.19 GPa using aqueous suspensions of partially oxidized black phosphorus (oBP) nanosheets. Comprehensive experimental characterization combined with theoretical modeling revealed that the abundant surface P=O and P-OH functional groups on oBP nanosheets played a dual role: (1) they effectively anchored water molecules at the tribological interface even under ultrahigh pressure conditions, and (2) mediated the transformation of the contact interface from direct solid-solid interaction to a more favorable oBP-oBP sliding configuration. This interfacial restructuring mechanism, facilitated by the unique surface chemistry of oBP nanosheets, was identified as the key factor enabling the unprecedented pressure stability of the superlubricating state. In 2025, Li et al.[154] achieved rapid superlubrication (reaching $\mu \approx 0.0048$ in just 26 seconds under a contact pressure of 710 MPa) on engineered steel surfaces by introducing phytic acid (PA)-modified BP nanosheets (PA-BP) into a poly(aspartic acid) (PASP) and ethylene glycol (EG) solution (PASP/EG). The addition of PA-BP reduced the wear rate by 96%

compared to PASP/EG alone. Surface analysis and molecular dynamics simulations revealed that PA-BP nanosheets readily adsorbed onto the steel surface, forming a tribofilm. During friction, these nanosheets underwent oxidation, enabling them to adsorb water molecules from PASP/EG and create a stable 18 Å-thick water layer. This transformed the shear interface into a low-strength water layer, facilitating rapid superlubrication even at extreme pressures.

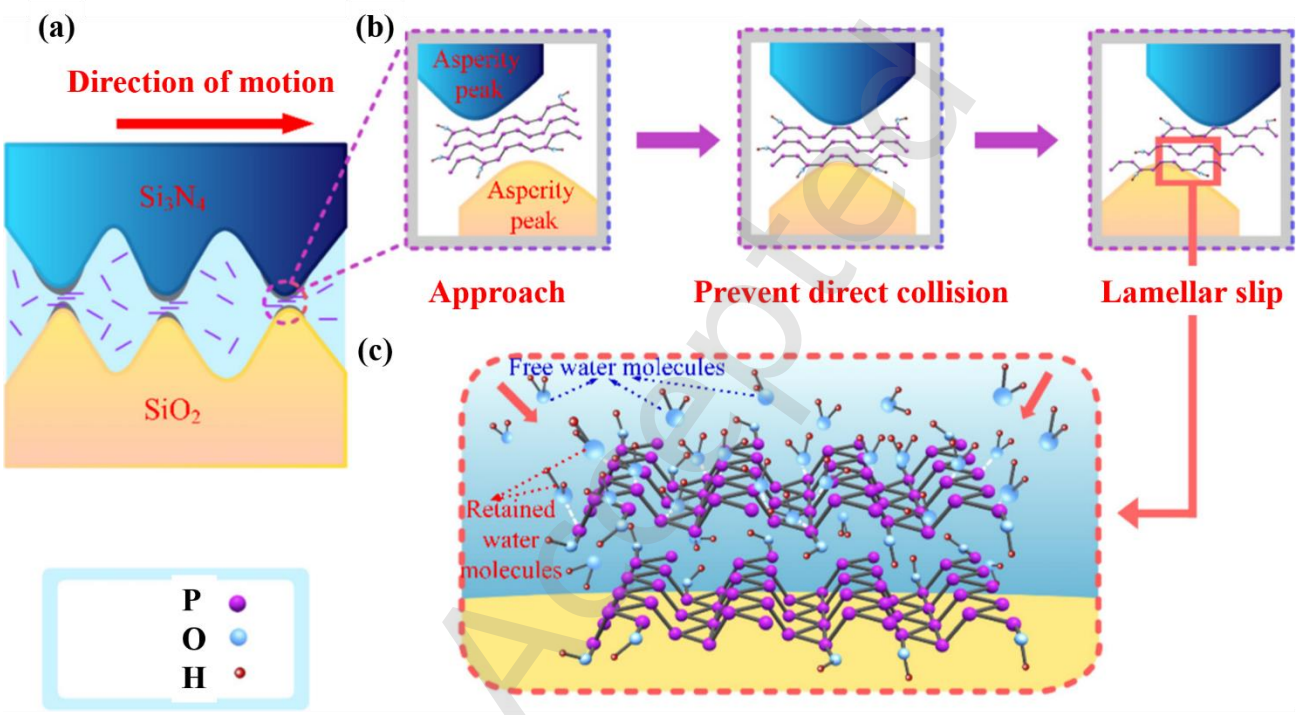

Fig. 8 (a) Schematic illustration of the sliding interface lubricated by BP-OH nanosheets. (b) Proposed dynamic lubrication mechanism of the BP-OH nanosheets. (c) Schematic depicting a stable water layer retained near the hydrophilic BP-OH nanosheet surface, resulting in low friction. Reprinted from ref [151] with permission from ACS Publications, Copyright 2018.

While conventional polytetrafluoroethylene (PTFE) coatings can form transfer films at tribological interfaces to achieve low friction coefficients (typically 0.05-0.15), their practical application is limited by high wear rates resulting from poor interfacial adhesion and inadequate mechanical strength. The incorporation of black phosphorus (BP) nanosheets into PTFE matrices demonstrates remarkable improvements: BP/PTFE composite coatings exhibit a 61% reduction in friction coefficient compared to pure PTFE coatings. Microstructural analysis reveals that BP forms a composite lubricating film at

the contact interface, which effectively mitigates mechanical wear induced by adhesive interactions between sliding surfaces. This enhancement stems from BP's unique nano-pleated structure, which exhibits a negative Poisson's ratio ($\nu \approx -0.027$). This auxetic behavior causes lateral expansion under tensile stress and transverse contraction under compression within the elastic regime. During frictional contact, BP layers undergo lateral expansion that dissipates energy through elastic deformation, thereby reducing both groove formation and adhesive wear. Furthermore, the weak van der Waals interactions between adjacent BP nanosheets in the tribofilm facilitate interlayer sliding, significantly decreasing shear resistance. This dual mechanism - energy absorption through auxetic deformation and reduced interfacial shear through lamellar sliding - collectively enhances the coating's anti-friction performance and durability, as evidenced by extended service life in tribological testing[140]. Lv et al.[155] conducted a systematic investigation on the tribological performance of polyetheretherketone (PEEK)/polytetrafluoroethylene (PTFE) and carbon fiber (CF)/PTFE composites reinforced with BP nanosheets. Experimental results demonstrated a remarkable reduction in the coefficients of friction (COFs) for both composite systems upon BP nanosheet incorporation, achieving a minimum COF value of 0.04 - representing a 75% reduction compared to pristine PTFE composites. Notably, the wear behavior exhibited material-dependent characteristics: the PTFE/PEEK composite showed significant wear resistance improvement, whereas the CF/PTFE composite displayed an inverse correlation between wear rate and filler concentration. Surface characterization through morphological and elemental distribution analyses revealed that BP nanosheets preferentially formed a continuous tribofilm consisting of phosphorus oxides and phosphoric acid at the contact interface, effectively replacing the conventional PTFE transfer film mechanism. This in situ generated BP-derived tribofilm was identified as the primary contributor to both friction reduction and the suppression of adhesive wear phenomena. In their 2022 study, Sun et al.[156] systematically examined the dry-sliding tribological performance and wear mechanisms of PEEK composites incorporating PTFE and BP nanosheets. The optimized PEEK/10 wt% PTFE/0.5 wt% BP composite demonstrated remarkable

improvements over pure PEEK, achieving a 73% reduction in coefficient of friction (from 0.369 to 0.097) and a 95% decrease in wear rate (from 1.0×10^{-4} $mm^3/N\cdot m$ to 5.1×10^{-6} $mm^3/N\cdot m$). These enhancements were attributed to the formation of a protective BP-derived transfer film at the sliding interface. Beyond tribological benefits, the composite exhibited superior biological properties, including enhanced surface wettability and exceptional antibacterial performance (＞99.9% bacterial inhibition), positioning it as a promising candidate for biomedical implant applications. In their 2022 study, Wang et al.[157] successfully fabricated degraded black phosphorus/polytetrafluoroethylene (d-BP/PTFE) composites via spark plasma sintering (SPS). Tribological evaluation revealed that the composite containing 5 wt.% d-BP exhibited optimal friction-reducing and anti-wear performance in both dry sliding and aqueous environments. Surface characterization of counterpart balls and wear tracks identified the formation of a phosphorus oxide-rich transfer film as the primary mechanism responsible for the significant reduction in both coefficient of friction (COF) and wear rate. Notably, under aqueous conditions, the phosphorus oxide layer demonstrated additional lubrication enhancement through dissolution and subsequent formation of an interfacial hydrogen-bond network, which further minimized frictional resistance.

5 2D Carbon Materials in Tribology

Carbon-based structural materials have emerged as pivotal components in advanced lubrication systems[23,158]. Graphite, with its characteristic layered structure, demonstrates exceptional tribological properties through the formation of interlayer slip planes at friction interfaces. Its remarkable chemical stability ensures consistent lubrication performance even under elevated temperatures, and it also possesses inherent electrical conductivity, which makes it particularly suitable for applications in high-temperature bearings, sealing components, and conductive lubricant coatings. Another significant advancement comes from diamond-like carbon (DLC) films[159], which combine near-diamond hardness with outstanding wear resistance. The tunable nature of DLC films - achieved through precise control of sp^3/sp^2 hybridization ratios or strategic doping with metallic/light elements

- allows for customized optimization of mechanical properties (hardness and toughness) and frictional characteristics. This adaptability enables DLC films to satisfy the stringent requirements of aerospace applications and precision instrumentation, performing reliably across extreme conditions including vacuum, high-temperature (≤ 350 °C), and cryogenic environments. The following discussion will systematically examine recent tribological advancements in 2 D carbon structures, with particular focus on graphene and graphene derivatives including oxidized and fluorinated graphene variants.

5.1 Graphene

Graphene possesses a distinctive 2D hexagonal honeycomb lattice structure, where each monolayer consists of sp^2-hybridized carbon atoms interconnected through robust covalent bonds, endowing the material with exceptional in-plane mechanical strength. The interlayer interactions in multilayer graphene are governed by weak VDW forces, which facilitate relative shear motion between adjacent layers. This unique combination of strong intralayer bonding and weak interlayer coupling is fundamentally responsible for graphene's outstanding tribological properties, particularly its remarkable friction-reducing capabilities[160]. Both microscopic[161] and mesoscopic[49] investigations have demonstrated that the periodic variation of interlayer friction is intrinsically linked to the rotational symmetry characteristics of the atomic lattice. Remarkably, when the contacting layers adopt an incommensurable stacking configuration, this structural arrangement can lead to the emergence of superlubricity - a near-zero friction state. Ma et al.[46] achieved a remarkably stable ultra-low friction coefficient ($\mu \approx 0.003$) at the graphene/graphene interface by employing a graphene-coated SiO_2 probe. Their analysis revealed that the observed superlubricity stability stems from the formation of overall incommensurable contact configurations at the sliding interface. In 2024, Liu's research group[47] achieved a significantly extended superlubricity lifetime (> 1.5 hours) during the interlayer sliding of graphene through their novel nanomanipulation technique. Their combined experimental and theoretical investigations revealed that the interlayer friction of graphene stems from the dynamic formation of π-π stacking interactions during relative sliding motions, which creates

continuous potential energy wells. Lee et al.[162] characterized the nanoscale surface friction of graphene sheets deposited on silicon wafers, uncovering an intriguing thickness-dependent frictional behavior. Their results demonstrated a progressive decrease in friction with increasing layer number, ultimately converging to bulk-like friction values. This phenomenon was found to be independent of experimental parameters including normal load, sliding velocity, and tip material composition. They proposed a mechanistic explanation based on elastic deformation principles: During AFM tip scanning on weakly adhered 2D materials, the strong tip-sample adhesion induces out-of-plane wrinkling of the atomic sheets ahead of the scanning tip. The bending stiffness, which scales with thickness, plays a critical role in this process - thicker samples exhibit greater resistance to wrinkling, leading to reduced real contact area and consequently lower friction (Figure 9). To validate this mechanism, they performed control experiments using graphene deposited on high-viscosity mica substrates, where the constrained out-of-plane deformation effectively eliminated the thickness-dependent friction behavior. Subsequent experimental and theoretical studies have consistently reproduced the thickness-dependent wrinkling phenomena, establishing this as a universal characteristic of 2D materials on weakly bonded substrates[163]. Paolicelli et al.[164] systematically verified the inverse correlation between friction force and layer number across diverse environmental conditions (vacuum, dry nitrogen, and humid air), demonstrating the robustness of this phenomenon. Through molecular dynamics (MD) simulations of diamond tip-graphene interactions, Ye et al.[165] further elucidated the mechanistic relationship between frictional response and out-of-plane folding-induced wrinkles in suspended few-layer graphene. These findings align with and substantiate the earlier theoretical framework proposed by Mohammadi et al.[166] regarding thickness-dependent tribological behavior in 2D materials. In a recent study, Zhang et al.[167] demonstrated a solid-solid interfacial quantum friction phenomenon through AFM measurements on folded graphene. Their findings revealed that the frictional force at folded graphene edges exhibits a nonlinear increase with the number of layers, contrasting with the above theory. This anomalous behavior is principally explained by strain-induced pseudo-Landau

quantization: it selectively suppresses electronic energy dissipation at folded edges, yet phononic dissipation proceeds normally irrespective of folding. In their pioneering work, Chen et al.[168] elucidated the fundamental origins of friction through atomic-scale investigations of silicon probe sliding on graphite surfaces with atomic steps. Their comprehensive analysis revealed that frictional forces arise from synergistic contributions of: (1) physical interactions governed by surface morphological characteristics, and (2) chemical processes mediated by terminal functional groups. Based on this dual-mechanism framework, they proposed a friction modulation strategy that exploits the competitive interplay between these two dominant processes. Building upon their previous findings, Chen's group conducted an in-depth investigation into the physicochemical origins of adhesion and friction heterogeneity at atomic step edges[169]. Their analysis identified two distinct but interrelated mechanisms governing frictional behavior at graphene step edges: (1) a geometric component arising from topographical height variations, and (2) a chemical component mediated by tip-edge interactions. Computational simulations revealed that the geometric effect stems from elastic deformation of the AFM tip during scanning, producing resistance during ascending motion while generating an assistive force during descent. In contrast, the chemical effect was attributed to hydrogen bonding between the tip's hydroxyl groups and step edges, which consistently generated resistance regardless of scanning direction. This mechanistic separation explains the observed decoupling between measured friction and adhesion forces at step edges, challenging the applicability of conventional contact mechanics models for atomically precise surface features. Through systematic investigations of graphene step edges with controlled height but varying terminal chemistries, the research team uncovered critical interface-dependent friction mechanisms[170]. Their experimental design compared two distinct configurations: (1) hydroxyl-terminated edges under alcohol vapor exposure, where physical adsorption of alcohol molecules induced passivation of hydroxyl groups, and (2) encapsulated graphene configurations creating chemically inert reference structures. Combined experimental and computational analyses revealed hysteresis phenomena in bidirectional scanning friction

measurements across all chemically active edge types. This directional dependence was mechanistically linked to anisotropic deformation behavior of edge-terminating groups, demonstrating how molecular-scale structural asymmetries propagate to macroscopic tribological responses.

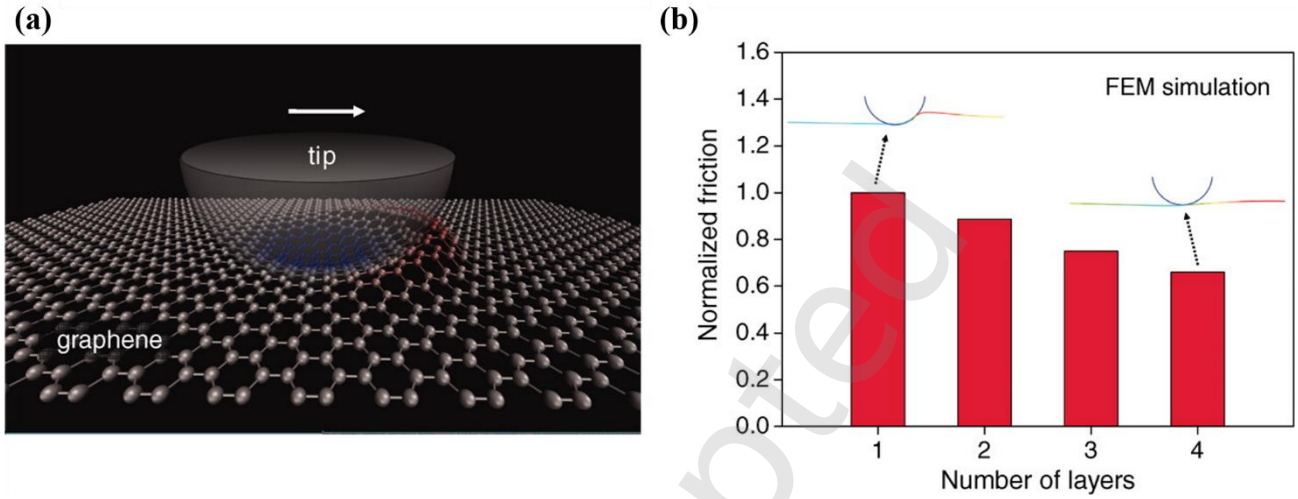

Fig. 9 (a) Schematic of the wrinkling effect; (b) Simulation results indicating friction decreases with increasing layer number. Reprinted from ref [162] with permission from AAAS, Copyright 2010.

Graphene has emerged as a pivotal material in recent years for enhancing lubricant performance, owing to its unique layered structure and exceptional mechanical properties that contribute to superior anti-friction and wear resistance characteristics. Nevertheless, the critical challenge persists in effectively translating these nano/micro-scale advantages into practical macro-scale lubrication applications. Recent investigations[171] have systematically evaluated the macroscopic tribological performance of graphene-enhanced PAO lubricants through comparative studies of steel/steel and DLC/DLC contact configurations. The incorporation of graphene nanoparticles demonstrates remarkable friction reduction effects, achieving approximately 44% and 50% reduction for steel and DLC contacts respectively under boundary lubrication regimes. This enhancement primarily stems from the in-situ formation of graphene-derived tribofilms at both steel and DLC-coated interfaces. Critical parameters governing tribofilm formation and morphology include: (1) nanoparticle concentration - higher graphene loading promotes more extensive tribofilm coverage as nanoparticles accumulate within surface asperities, though DLC surfaces require greater concentrations due to

weaker graphene adhesion; and (2) contact speed - effective tribofilm formation occurs predominantly under boundary lubrication (BL) conditions at lower speeds, while elastohydrodynamic lubrication (EHL) at higher speeds with thicker oil films shows negligible surface interaction. These findings elucidate the dual dependence of lubrication performance on additive concentration and operational parameters, providing fundamental insights into tribofilm formation mechanisms[172,173]. The in-situ generation of graphene films from lubricating oil has emerged as a highly promising research area. Recently, Li et al.[174] pioneered a novel approach by utilizing iron single atoms to catalyze a series of diesters, with dioctyl malate (DOM) as a representative example, for the production of graphitic tribofilms. These tribofilms demonstrated remarkable performance, reducing wear by factors of 5-7 and friction by over 50% compared to base oil under high-pressure conditions. The iron single atoms are generated in-situ through mechanochemical effects during sliding contact. Notably, the 30 nm graphitic tribofilms preferentially form from diesters derived from short-chain carboxylic acids due to their lone pair effect, which effectively stabilizes carbon free radicals. Computational simulations have further elucidated the intricate steps involved in the catalytic decomposition of DOM by iron and the subsequent formation of the graphitic carbon tribofilm.

Extensive research has demonstrated the exceptional tribological performance of graphene coatings. Notably, Berman et al.[175] realized macroscale superlubricity at DLC-SiO_2 interfaces through an innovative architecture where graphene nanosheets encapsulate nanodiamonds to form stable nanoscrolls. Parallel advancements by the Androulidakis[176] and Li[177,178] research groups have independently achieved interlayer superlubricity in graphene coatings via controlled in-plane strain engineering approaches. Graphene nanosheets have demonstrated remarkable potential as reinforcement phases in metal matrix composites, substantially enhancing both mechanical and tribological characteristics[179,180]. In a notable study by Xue et al.[181], the incorporation of graphene nanosheets into TiAl-based composites was shown to significantly improve their mechanical properties and tribological performance under dry sliding conditions. The research revealed that adding

merely 3 wt% graphene nanosheets to the titanium alloy matrix yielded extraordinary property enhancements: 129% improvement in microhardness, 149% increase in fracture toughness, along with 37% reduction in friction coefficient and 78% decrease in wear rate. These dramatic improvements are attributed to the effective load transfer and self-lubricating properties imparted by the graphene reinforcement. Hsieh et al.[182] reported substantial improvements in both mechanical properties (including Vickers hardness and compressive yield strength) and tribological performance in aluminum alloys reinforced with merely 0.25 wt% graphene. Furthermore, research has documented the significant optimization effect of graphene nano-adsorbents on the comprehensive mechanical and tribological characteristics of Inconel 718-based composites[183]. Multiple factors critically influence the mechanical and tribological properties of graphene-reinforced metal composites. The graphene content and operational environment play pivotal roles in determining composite performance. Beyond optimal external conditions (including applied load, sliding speed, and reinforcement content), several intrinsic factors contribute to property enhancement: (1) the superior strength and homogeneous dispersion of graphene additives, (2) induced grain refinement, and (3) increased dislocation density within the matrix. These microstructural modifications synergistically improve both bulk mechanical properties and localized anti-friction/anti-wear characteristics. Furthermore, the remarkable tribological improvement stems from the in-situ formation of protective tribofilms at contact interfaces. These self-generated films effectively isolate mating surfaces, minimizing direct asperity contact and consequently reducing both frictional resistance and material loss rates. The enhanced tribological performance of graphene nanosheet (GNP)-reinforced ceramic composite coatings primarily stems from two synergistic mechanisms: (1) the intrinsic toughening effects of GNPs including interlayer sliding, crack bridging, and crack deflection; and (2) their exceptional solid lubrication properties[184]. A representative case study demonstrates this enhancement in GNPs/Al_2O_3-TiO_2 composite coatings, where fracture toughness exhibits a positive correlation with the elastic modulus-to-hardness ratio (E/H). Specifically, with the incorporation of 1.5 wt% GNPs, the elastic modulus increases

dramatically from 60.4 GPa to 102.31 GPa, while hardness improves from 7.54 GPa to 10.31 GPa. This substantial increase in the E/H ratio directly contributes to superior fracture toughness in the composite coating. The improved mechanical properties effectively mitigate abrasive particle grooving and crack propagation during wear processes, consequently achieving optimal tribological performance characterized by reduced friction coefficients and enhanced wear resistance[185].

5.2 Oxidized/Fluorinated Graphene

Both oxidized and fluorinated graphene derivatives retain the fundamental layered architecture characteristic of pristine graphene. However, their surface chemistries exhibit distinct differences: (1) Pristine graphene features a passivated surface with edge-terminated hydroxyl groups (-OH) as the primary functional moieties; (2) Oxidized graphene demonstrates significantly enhanced surface reactivity through abundant oxygen-containing functional groups (-O-, -OH, -COOH) distributed across its basal planes; (3) Fluorinated graphene presents fluorine termination (-F) predominantly at its edges, creating a unique surface electronic structure while maintaining the layered framework. Experimental investigations reveal that surface chemical functionalization significantly alters nanoscale frictional behavior[186]. While fluorinated graphene exhibits reduced adhesion forces with AFM tips compared to pristine graphene, lateral force measurements demonstrate a sixfold increase in friction. This apparent contradiction can be explained through DFT calculations, which indicate that fluorination enhances the bending stiffness of graphene. The increased rigidity creates additional energy dissipation during tip movement, as the AFM probe must overcome greater resistance when traversing the deformed fluorinated graphene surface under applied normal loads. Through systematic AFM investigations, Zeng et al.[187] elucidated the distinct tribological behaviors of pristine graphene (PG), oxidized graphene (GO), and fluorinated graphene (FG). Their comparative analysis revealed fundamentally different load-dependent friction responses: GO with strong surface adhesion demonstrated nonlinear friction-load dependence, whereas PG and FG with weaker adhesion exhibited linear relationships. Notably, the study uncovered a dynamic adhesion enhancement phenomenon post-

sliding, where both the adhesion enhancement magnitude and transient friction increase correlated positively with surface energy. This synergistic effect stems from the interplay between tip sliding dynamics and surface hydrophilicity. Atomic-scale observations further verified the sliding-induced interface interaction enhancement, with the strengthening degree being directly proportional to the surface's hydrophilic character.

Graphene oxide (GO) materials, encompassing both pristine and functionalized forms, exhibit wide-ranging applications as lubricant additives[188,189]. Their anti-wear mechanism primarily operates through the formation of protective GO adsorption layers and tribochemical films, which effectively isolate friction pairs and minimize direct contact, consequently reducing wear. In their 2016 study, Chen et al.[190] conducted a systematic investigation of few-layer graphene oxide (GO) sheets as additives in hydrocarbon base oils. Their research revealed that the incorporation of GO sheets led to simultaneous reductions in both the coefficient of friction (COF) and wear rate, while also extending the effective working temperature range of the lubricant. The researchers proposed that this enhanced tribological performance stems from the GO sheets' ability to adhere to sliding surfaces during friction processes. These adhered nanosheets function as protective interfacial films, effectively preventing direct contact between the mating surfaces and thereby reducing both friction and wear. Wu et al.[191] systematically investigated the tribological performance of oxidized graphene nanosheets (GOPs) as lubricant additives in Si_3N_4 ceramic/GCr15 steel sliding pairs under varying rotational speeds and applied loads. Their experimental data demonstrated significant improvements with 0.5 wt% GOPs addition: 18.2% enhancement in load-bearing capacity, 14.7% reduction in friction coefficient, and 34% improvement in wear resistance. The characterization results revealed that the GOPs effectively migrated to the contact interface, forming a protective boundary layer that minimized direct asperity contact between mating surfaces, thereby suppressing material transfer and substantially reducing both friction and wear. Fan et al.[192] developed a novel oxygen/fluorine dual-functionalized graphene (OFG) through selective fluorination of graphene oxide (GO) while preserving its layered structure.

When employed as a water-based lubricant additive, OFG demonstrated superior wear resistance, exhibiting 47% and 31% lower wear rates compared to pure water and conventional GO additives, respectively. The enhanced performance stems from OFG's unique dual functionality: the abundant oxygen-containing groups improve aqueous dispersibility through enhanced hydrophilicity, while the incorporated fluorine atoms contribute to exceptional tribological properties via intrinsic self-lubrication mechanisms at the friction interface. Zhang et al.[193] developed two novel graphene-based lubricant additives (GO-D and GO-T) through covalent functionalization of carboxyl groups of graphene oxide (GO) with 1-decylthiol and tertiary-decylthiol, respectively. Comparative evaluation of their dispersion stability and tribological properties in rapeseed oil revealed that GO-D exhibited superior performance to both GO-T and pristine GO. Specifically, GO-D demonstrated enhanced colloidal stability and more effective friction/wear reduction. This performance enhancement primarily originates from the formation of robust adsorption layers and tribochemical films at the friction interface, with the alkyl chain structure of GO-D proving particularly effective in surface protection. Above research have demonstrated that graphene oxide (GO) as a lubricant additive typically exhibits significantly higher macroscopic friction coefficients compared to superlubricity state. Ge et al.[194] proposed an innovative solution by synergistically combining graphene oxide nanoflakes (GONFs) with ethylene glycol (EDO) at the Si_3N_4-SiO_2 interface, achieving remarkable macroscopic superlubricity ($\mu = 0.0037$) coupled with ultralow wear. Their systematic investigation revealed a dual lubrication mechanism: (1) GONFs adsorbed onto friction surfaces effectively prevented direct asperity contact, while the resulting extremely low interfacial shear stress promoted superlubricity; (2) Concurrently, hydrogen-bond-mediated formation of hydrated GONFs-EDO networks established partial slip boundary conditions, creating a fluid lubrication film with minimal shear resistance (Figure 10). This breakthrough demonstrates the potential of solid-liquid coupling lubrication systems for achieving simultaneous superlubricity and wear protection, offering new design principles for advanced lubrication technologies. Subsequently, Ge et al.[195] demonstrated a robust macroscale

liquid-superlubricity state (μ ≈ 0.005) under an extreme pressure of 600 MPa, achieved by combining GO nanosheets with an ionic liquid (IL) between the friction pairs of Si_3N_4/sapphire. Analysis revealed that a composite boundary layer formed by IL at the interface significantly enhanced anti-wear performance, while GO nanosheets were observed to adsorb directly onto worn surfaces, effectively transforming the shear interface from Si_3N_4/sapphire into GO/GO. This transition, coupled with the extreme pressure resistance and ultralow interlayer shear stress of GO nanosheets, enabled the superlubricity state. Thus, the synergistic interplay between GO nanosheets and IL played a pivotal role in achieving macroscale liquid-superlubricity under extreme pressure.

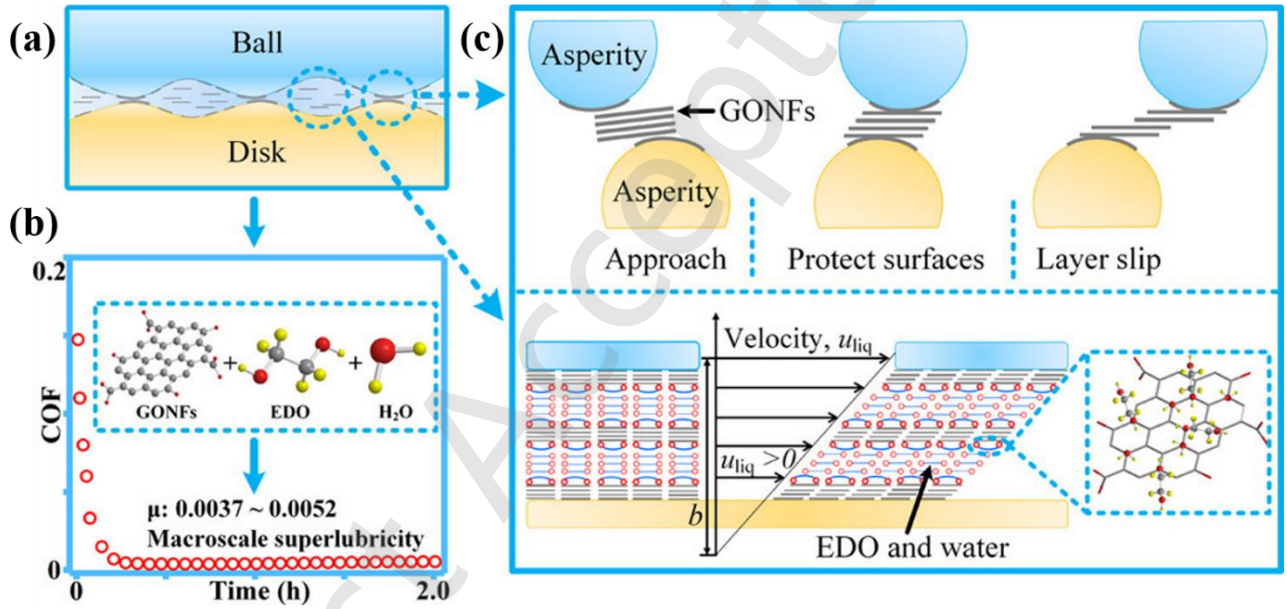

Fig. 10 (a) Schematic of the GONFs-EDO system, (b) its lubrication performance, and (c) friction-reduction mechanism. Reprinted from ref [194] with permission from ACS publications, Copyright 2018.

The reduced graphene oxide (rGO) coating demonstrates remarkable potential for friction and wear reduction. According to Kim et al.[196], applying rGO coating to 440C stainless steel balls resulted in a sixfold decrease in friction between the balls and plate under water lubrication conditions. Notably, the friction coefficient of rGO-coated balls with water lubrication (~0.1) matched that of uncoated balls with oil lubrication. Furthermore, the wear rate of steel plates sliding against rGO-coated balls under water lubrication (1×10^{-10} mm^3/Nm) was three times lower than that observed with uncoated

balls under oil lubrication. The researchers hypothesized that water molecule penetration into the rGO layer contributed to the coating's reduced shear strength, explaining these significant improvements in tribological performance. Mo et al.[197] developed a novel composite coating on silicon substrates by combining multiple alkylated cyclopentane (MAC) with reduced graphene oxide (rGO), where the rGO was synthesized from expandable graphite powder. The study revealed that this composite coating exhibited excellent mechanical properties, including high load-bearing capacity, strong adhesion, and superior tribological characteristics. Specifically, the coating demonstrated outstanding anti-wear properties and achieved an exceptionally low friction coefficient ranging from 0.02 to 0.03. The researchers attributed these enhanced performance characteristics to the functionalized graphene oxide, which served as an effective reinforcing phase in the coating. Zhao et al.[198] developed an innovative approach by in situ functionalizing graphene oxide (GO) with ionic liquid (IL) and subsequently dispersing it uniformly within an epoxy resin (EP) matrix. Their research demonstrated that the IL-functionalized graphene oxide (GO-IL) exhibited significantly better dispersion in the EP matrix compared to freshly exfoliated GO. Tribological testing revealed that even at low concentrations, GO-IL additives dramatically reduced both friction and wear in the EP composite, with particularly notable performance enhancements under severe sliding conditions. This improvement was attributed to a distinct synergistic effect between the IL and GO components. Advanced interface characterization techniques confirmed the formation of a unique nanostructured tribofilm consisting of GO nanosheets, IL functional groups, and EP matrix components. The researchers proposed that the GO-IL complex facilitated sustained release of EP molecules, which served as building blocks for tribofilm formation, while the graphene oxide component simultaneously strengthened the film's mechanical integrity and enhanced its boundary lubrication properties.

6 2D Organic Materials in Tribology

The application of organic lubricating materials dates back centuries, with initial developments primarily concentrating on three-dimensional polymer-based self-lubricating friction pairs[26,199].

The inherent compliance of organic materials and the rotational freedom of C-C bonds within their molecular structures initially hindered the development of 2D organic materials, resulting in delayed research progress on their lubrication properties. Consequently, investigations into 2D organic lubricants remain at an early stage of exploration. This emerging field originated from studies on organic molecular assembly structures and has recently gained significant momentum through advancements in covalent organic frameworks (COFs), which have opened new avenues for lubrication research.

6.1 Organic Molecular Assembly Structures

Self-assembly describes the spontaneous organization of fundamental structural units into stable, ordered architectures through non-covalent interactions. In organic molecular self-assembly systems, the basic building blocks are organic molecules that interact primarily via VDW forces or hydrogen bonding. The formation of 2D assemblies frequently exploits interface confinement effects, particularly at solid-liquid interfaces. This section examines three principal types of organic molecular assembly architectures: single-component networks, multi-component networks, and host-guest assembly structures.

In 2017, Shi et al.[200] investigated the supramolecular assembly of sugar acid molecules on pyrolytic graphite surfaces via hydrogen bonding and conducted nanoscale friction measurements. Their findings revealed that the introduction of pyridine molecules as structural regulators significantly increased the frictional force. This effect was attributed to enhanced probe-molecule interactions, which raised the energy barrier required for probe sliding. In 2018, Shi et al.[36] constructed seven distinct supramolecular networks on highly oriented pyrolytic graphite (HOPG) through hydrogen bonding and van der Waals (VDW) interactions. Their combined experimental and theoretical analyses revealed a direct correlation between frictional energy dissipation and both intermolecular interaction energies and molecule-substrate interfacial interactions (Figure 11). Tan et al.[201] demonstrated nanoscale superlubricity in host-guest assembly structures comprising fullerene derivatives and

macrocyclic templates on HOPG. Furthermore, they established that an external electric field could precisely modulate these systems' tribological properties[202]. In 2021, Tan et al.[203] successfully fabricated self-assembled liquid crystal molecular films on pyrolytic graphite substrates, achieving microscale superlubricity through AFM probe sliding experiments[204]. Their DFT calculations qualitatively established the correlation between the friction coefficient and interfacial interaction energies. Tan et al.[205] fabricated hydrogen-bonded self-assembled monolayers of uracil-functionalized boron dipyrromethene (BODIPY) derivative on pyrolytic graphite. Nanotribological characterization revealed that the hydrogen-bonded network enhanced probe-molecule interactions, leading to increased frictional forces. In 2023, Qiao et al.[206] systematically fabricated three distinct phthalocyanine self-assembled monolayers (SAMs) with controlled molecular architectures: planar-oriented, edge-oriented, and hierarchical multi-level structures. Through comprehensive nanotribological characterization, they elucidated the structure-property relationships governing the frictional performance of these boundary lubricants. The study revealed that planar-oriented SAMs exhibited superior tribological properties compared to their edge-oriented counterparts. Notably, the hierarchical architecture demonstrated anisotropic mechanical responses under shear stress, with direction-dependent deformation resistance arising from structural ordering and stiffness anisotropy, ultimately leading to multistate frictional behavior.

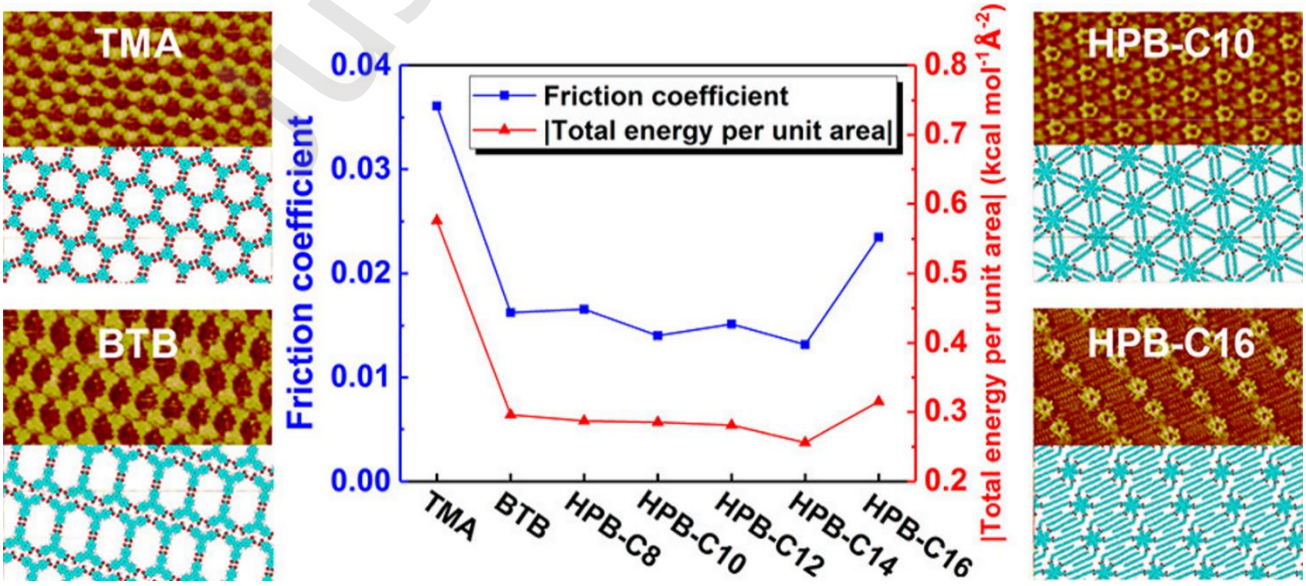

Fig. 11 Friction coefficient versus total energy per unit area for different molecular self-assembly structures. Reprinted from ref [36] with permission from ACS publications, Copyright 2018.

While solvent-induced dissociation of organic molecular assemblies often limits their application as lubricant additives, recent studies demonstrate that lubricant molecules can undergo pressure-induced phase transitions at contact interfaces, facilitating in situ formation of self-assembled monolayer films on friction surfaces. Jin et al.[207] successfully engineered an in-situ heterojunction through the interfacial coupling of a crystalline boundary tribofilm with a pressure-induced solid-phase 1-dodecanol molecular layer, thereby realizing structural superlubricity at the liquid-solid interface. Both AFM characterization and MD simulations consistently revealed a 180-degree periodic friction behavior in this heterojunction system. Particularly noteworthy is the emergence of phase transition structural superlubricity (PTSS), which occurs specifically when the molecular axis of 1-dodecanol adopts a perpendicular orientation (90°) relative to the sliding direction. Considering that the PTSS state in the aforementioned system was initially limited to a maximum Hertz contact stress of 600 MPa, the researchers subsequently developed an advanced approach by covalently functionalizing graphene with polydopamine[208]. This surface modification substantially improved the adsorption capability between graphene and SiC substrate, facilitating the formation of a well-defined crystalline boundary tribofilm with a controlled thickness ranging from 180 to 200 nm. Through the strategic construction of an in-situ heterojunction between this graphene-based tribofilm and the solid-phase 1-dodecanol molecular layer, the team successfully extended the PTSS regime to withstand Hertz contact stresses as high as 1.313 GPa at the liquid-solid interface. It should be noted that 1-dodecanol undergoes a distinct liquid-to-solid phase transition when subjected to contact stresses exceeding 137 MPa. In this solid phase, the molecules spontaneously organize into a characteristic herringbone pattern through intermolecular hydrogen bonding between hydroxyl groups.

6.2 Covalent Organic Frameworks

Covalent Organic Frameworks (COFs) represent a class of artificially synthetic porous crystalline materials constructed from precisely designed organic building blocks via robust covalent linkages[209]. These materials possess well-defined periodic architectures that endow them with exceptional physicochemical property, such as permanent porosity, remarkably high surface areas, outstanding thermal/chemical stability, structural diversity, and ultralow density[210]. Owing to these unique properties, COFs have attracted considerable research interest across multiple disciplines, with emerging applications in the field of tribology[18].

Employing dynamic covalent chemistry strategies, Feng et al.[211] achieved in 2021 the successful synthesis of continuous, structurally robust glassy COFs exhibiting exceptionally low defect densities. This breakthrough was accomplished through the judicious selection of 5-((4-formylphenyl)ethylene)-isophthalaldehyde (TFPA) and 5'-(4-aminophenyl)-[1,1':3',1''-terphenyl]-4,4''-diamine (TAM) as the fundamental building blocks for COF construction. The resulting amorphous COF demonstrated significantly reduced friction force characterized by AFM when compared to its crystalline counterparts, which was primarily ascribed to its minimized defect concentration. The research team identified the elimination of grain boundaries as the key structural feature responsible for the enhanced tribological performance of amorphous COFs. A systematic investigation of the nanotribological properties of honeycomb-structured COFs was conducted by Tan et al.[212] in 2022. These framework materials were synthesized through the Schiff base condensation between aldehyde and amine precursors and demonstrated a distinct positive correlation between their friction coefficients and pore dimensions (Figure 12). Notably, the formation of co-assembled complexes with coronene guest molecules resulted in significantly enhanced friction coefficients, which was mechanistically attributed to the introduction of additional energy barriers for interfacial slip by the incorporated guest species. In 2024, Liu and colleagues[47] successfully fabricated a porphyrin-incorporated tetragonal COF via liquid-liquid interfacial confinement strategy. Remarkably, the research team not only realized interlayer superlubricity in this COF system using their proprietary nanomanipulation technique, but

also elucidated the underlying friction mechanism through comprehensive theoretical simulations. Their findings revealed that the interlayer sliding dynamics are governed by the formation of transient π-π stacking interactions between aromatic moieties (including both porphyrin macrocycles and benzene rings), which generates periodic potential wells and consequently gives rise to frictional resistance.

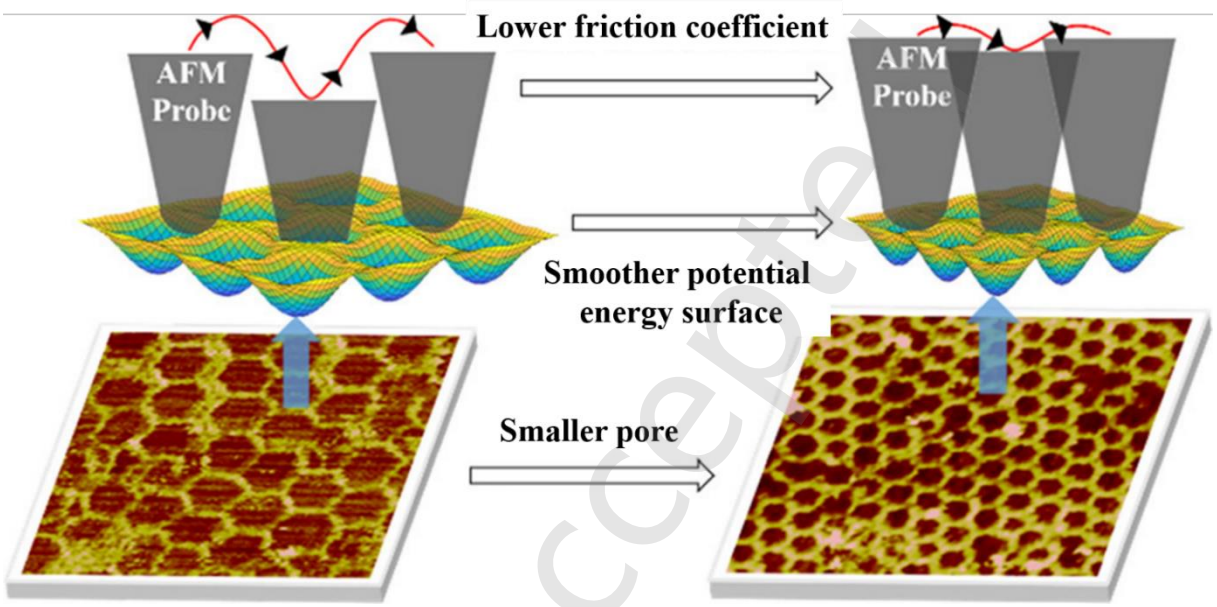

Fig. 12 Relationship between COF pore size, potential energy surface and surface friction. Reprinted from ref [212] with permission from ACS publications, Copyright 2022.

In their pioneering research, Wen and colleagues[213] introduced the 2D triazine-based COFs (TriCF) as lubricant additives in polyalphaolefin (PAO 10) base oil. Remarkably, the addition of 0.6 wt% TriCF nanoparticles to PAO 10 led to a dramatic reduction in friction coefficient (89.3%) and wear volume (77.8%) in the steel ball-on-steel disk contact. Through systematic characterization, they demonstrated that the unpaired electrons of nitrogen atoms in the triazine rings form strong charge-transfer complexes with the delocalized electrons on metal surfaces, thereby generating an ultrathin protective film that effectively minimizes direct asperity contact during sliding. Building upon their previous work, the research team successfully developed graphene-like COFs (GCFs) with triazine ring structures via an optimized ultrasonic-assisted solvothermal synthesis route[214]. AFM characterization revealed these GCFs possessed an ultrathin layered structure with a uniform thickness

of 3.1-3.3 nm. When incorporated as lubricant additives in PAO 10 base oil, the GCFs demonstrated exceptional colloidal stability and dispersion characteristics. Tribological characterization revealed that the GCF-modified lubricant exhibited exceptional performance at an ultralow concentration of 0.008 wt%, achieving a remarkable 53.5% reduction in friction coefficient and 95.4% decrease in wear volume. This remarkable performance was attributed to the unique p-d orbital hybridization between the electron-rich triazine moieties in GCFs and transition metal ions on the rubbing surfaces, which facilitated the formation of a robust, self-replenishing adsorbed film that effectively mitigated both mechanical wear and thermal degradation during sliding contact. In their subsequent investigation, Wen et al.[215] systematically explored the potential of COFs as high-performance water-based lubricant additives. Their study focused on 2D triazine-based COFs (2D-TriCF), which demonstrated excellent aqueous dispersibility through extensive hydrogen bonding interactions with water molecules. Tribological evaluations demonstrated that the addition of 0.1 wt% TriC to 1% aqueous solutions of coconut diethanol amide (CDEA) and Tween 85 (T-85) resulted in a significant reduction in both friction coefficient (72.2% and 74.4%, respectively) and wear rate (98.0% and 95.8%, respectively) compared to pure water. Surprisingly, the incorporation of 2D-TriCF additives led to a substantial mitigation of corrosion-induced wear. The exceptional anti-corrosion performance was ascribed to the synergistic interaction between TriC and surfactants. Specifically, the electron-rich triazine moieties in the 2D-TriCF framework formed coordination bonds with metal surfaces via lone pair electron donation, creating a robust interfacial protective layer. This molecular-level foundation further promoted the hierarchical self-organization of CDEA and T-85 surfactants into well-ordered micellar architectures. COFs can acquire enhanced or novel functionalities through post-synthetic modification strategies. In a notable advancement, Liu et al.[216] incorporated the commercial additive di-alkyl disulfonate (DDP) into 2D COFs to optimize their tribological performance. The DDP-functionalized COFs (DDP@TD-COF) were synthesized via a UV-triggered thiol-ene "click" reaction between vinyl-modified 2D COFs (TD-COF) and thiol moieties of DDP molecules, as illustrated in

Figure 13(a). The resulting nanosheets exhibited a uniform thickness of 3-4 nm. Remarkably, when dispersed in 500-SN base oil, DDP@TD-COF demonstrated exceptional lubricating efficacy, achieving a 50% reduction in friction coefficient and a 95% decrease in wear volume compared to untreated oil (Figure 13b). Mechanistic analysis revealed that the DDP@TD-COF nanosheets penetrate interfacial contact zones during sliding motion, forming durable lubrication films through synergistic interactions between their triazine components and metallic surface delocalized electrons (Figure 13c). Furthermore, the hierarchical COF architecture enables effective surface defect remediation through interlayer sliding mechanisms, whereby stacked nanosheets physically fill surface micro-pits while facilitating stress dissipation, collectively contributing to the observed friction and wear reduction. In 2024, Fan et al.[217] engineered three novel covalent-organic framework nanomaterials (CONs) featuring imide-bridged donor-acceptor architectures, demonstrating exceptional tribological performance in PEG 400 base oil. Advanced surface characterization revealed a groundbreaking mechanism where energy band matching between CONs and substrate species forms a Z-scheme heterojunction under friction, generating an internal electric field that traps ionic species within CON pores (pinning effect) while simultaneously attracting free CONs to assemble a secondary shear layer. This creates a unique dual-layered tribofilm with a chemically stabilized bottom layer bonded to the substrate and a mechanically adaptive top layer enabling easy shear. The synergistic combination of electrostatic modulation and layered architecture achieves remarkable friction reduction and anti-wear performance, establishing a new paradigm for smart lubricant design through nanostructure-substrate interaction engineering.

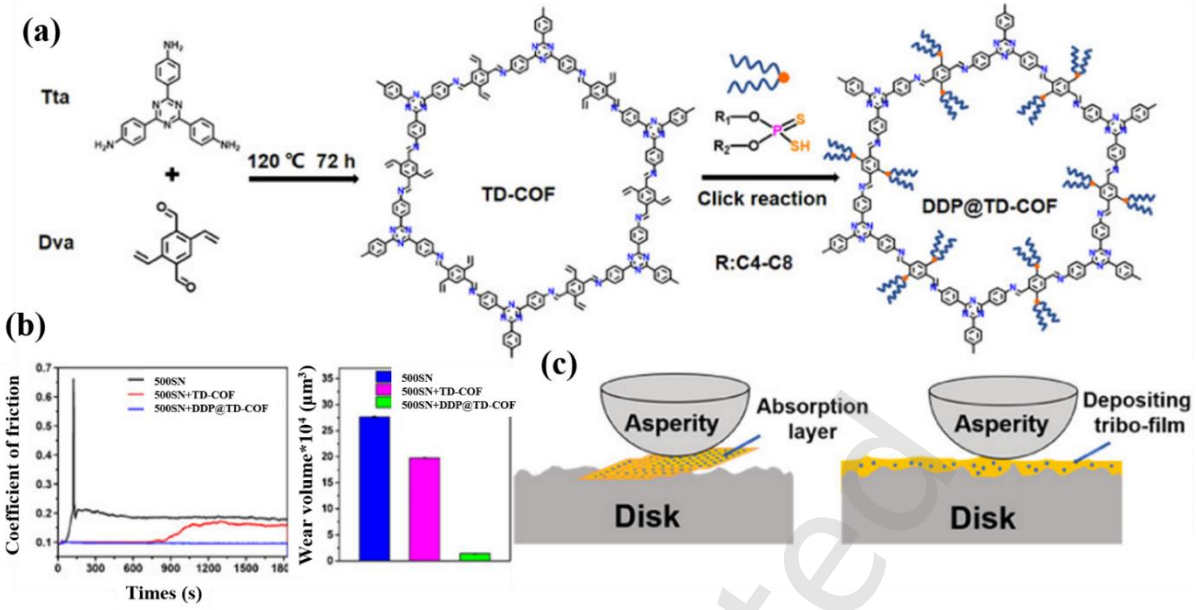

Fig. 13 DDP@TD-COF: (a) Synthesis, (b) tribological performance, and (c) friction-reduction mechanism in lubricant additives application. Reprinted from ref [216] with permission from ACS publications, Copyright 2021.

Self-lubricating composite materials (SLCs) represent a critical solution for extreme operating environments where traditional liquid lubricants prove ineffective. These advanced materials typically incorporate multiple reinforcing phases, with the integration of solid lubricants emerging as a particularly effective approach for friction and wear reduction. This strategy has demonstrated significant improvements in the tribological performance of various matrices, including metals, polymers, and ceramics. In a significant development, Liu et al.[218] successfully fabricated a graphene/COFs (G/COFs) hybrid composite via a one-step mechanochemical synthesis approach. When incorporated into bis-maleimide (BMI) resin, the resulting G/COFs/BMI composite exhibited remarkable tribological enhancements, showing 60% and 77.9% reductions in friction coefficient and wear rate, respectively, compared to pristine BMI. The lubrication mechanism involves the formation of a protective transfer film at the contact interface, which develops progressively as contact conditions deteriorate. This self-lubricating behavior, combined with the synergistic effects of graphene and COFs, contributes to the composite's outstanding performance. The COFs component not only

prevented the agglomeration of graphene nanosheets, but also improved the interfacial bonding between the filler and matrix, collectively resulting in optimized tribological properties. The application of COFs as reinforcing phases in composite coatings has been further demonstrated through their functionalization with siloxane polymers, significantly enhancing both corrosion resistance and tribological properties[219]. Polyhedral oligomeric silsesquioxane (POSS), a unique siloxane polymer, exhibits exceptional thermal and chemical stability owing to its inorganic Si-O-Si framework. POSS-modified polymers are particularly notable for their superior solubility, enhanced mechanical strength, excellent thermal stability, and remarkable corrosion resistance. Building upon these advantages, researchers successfully synthesized POSS-TpPa-1 composites via a Schiff base reaction between the carbonyl groups of 2D-COF (TpPa-1, Tp: 1,3,5-Triformylphrogroglucinol, Pa-1: 1,4-phenylenediamine) and the amine functionalities of POSS-NH_2 molecules. When incorporated into epoxy (EP) coatings, the resulting POSS-TpPa-1/EP composite demonstrated outstanding long-term anti-corrosion performance. This improvement stems from two key factors: (1) the weak interlayer interactions and high reactivity of TpPa-1 facilitate its homogeneous dispersion within the EP matrix, while (2) the synergistic interaction between POSS and TpPa-1 components simultaneously enhances both corrosion protection and wear resistance properties of the coating system.

7 2D Hybrid Materials in Tribology

Hybrid materials represent a distinct class of materials characterized by periodic structures comprising two or more components with distinct physico-chemical property. It is crucial to differentiate these from conventional composite materials: while hybrid materials exhibit long-range ordered crystalline structures without internal phase boundaries, composites consist of physically blended constituent materials that retain their separate phase identities through interfacial interactions. The fundamental distinction lies in their structural organization - hybrid materials demonstrate atomic-level periodicity throughout the entire structure, whereas composites display macroscopic phase separation with multiple interfaces. A representative case is the inorganic-organic hybrid structure, which constitutes

a crystalline material characterized by the ordered, periodic assembly of both inorganic and organic building blocks. Metal-organic frameworks (MOFs) serve as a quintessential manifestation of this structural paradigm, where inorganic nodes and organic linkers form well-defined, extended crystalline networks through coordination bonds[220]. Within the classification of 2D hybrid structures, we systematically incorporate 2D material heterojunctions as a distinct subclass. These heterostructures exhibit pronounced structural variations between their constituent layers, representing an alternative hybridization paradigm characterized by vertical (z-axis) integration of dissimilar materials.

7.1 Metal-Organic Frameworks

2D inorganic-organic hybrid materials have emerged as a research frontier in superlubrication fields, with metal-organic frameworks (MOFs) representing a prototypical example of such architectures[19]. These crystalline materials are formed through the spontaneous self-assembly of inorganic nodes (metal ions or clusters) and organic linkers (Polydentate molecular ligands), exhibiting unique mechanical properties that transcend conventional soft/hard material classifications[221]. Their well-defined crystalline structures feature: (1) compositional diversity, (2) programmable topological configurations, and (3) precisely tunable mechanical characteristics through molecular engineering. The exceptional structural predictability and design flexibility of MOFs enable systematic property modulation at the atomic level, making them particularly promising for developing next-generation multifunctional superlubricating materials with tailored performance characteristics.

In 2021, Liu et al.[222] constructed five square lattice MOFs sharing an identical crystal structure but differing in composition. Using AFM sliding measurements on their surfaces, these MOFs demonstrated excellent superlubricating properties, achieving an ultra-low friction coefficient of 5.3×10^{-4}. Through DFT simulations, they confirmed the microscopic friction mechanism involving an anchoring effect and established its relationship with the structural coordination stability. Finally, they proposed superlubricating structural design strategies based on crystal field theory (CFT) (Figure 14). Although surface structure undeniably governs friction behavior, the precise mechanisms mediating

this relationship await further clarification. In 2022, Liu et al.[39] engineered four distinct surface architectures in MOFs through surfactant-mediated modification. Atomic force microscopy-based tribological measurements revealed a remarkable structure-dependent friction modulation, with coefficients tunable across a wide range (5.6×10^{-4} to 2.1×10^{-3}). Through systematic analysis of atomic-scale friction force mapping and stick-slip dynamics, the team identified three molecular-scale friction mechanisms induced by surface functionalization: (i) molecular chain entanglement, (ii) viscoelastic hysteresis, and (iii) long-range adhesive interactions. Furthermore, their work established the beneficial role of surface porosity in friction reduction, providing new insights for designing low-friction MOF interfaces. Concurrently, Liu et al.[223] investigated the interlayer sliding behavior of tetragonal lattice MOFs in both homojunctions and heterojunctions using their proprietary developed technology. Their study revealed a counterintuitive finding that contrasts with conventional materials: homojunctions exhibit superior interlayer sliding capability compared to heterojunctions. Through subsequent DFT simulations, the researchers validated this conclusion and elucidated the underlying friction mechanism, attributing the sliding energy barrier to the vertical alignment of the congeneric components between layers. Furthermore, their work demonstrated that both the magnitude of friction resistance and the degree of friction anisotropy correlate with the coordination stability and lattice constant of the laminate structure. In their groundbreaking study[47], the research team investigated the structure-function relationship between structural factors and interlayer sliding behavior in 2D material homojunctions, particularly MOFs. Their work successfully identified two distinct interlayer friction mechanisms: potential barriers and potential wells. Beyond establishing the theoretical foundations for both interlayer friction and adhesion phenomena, the researchers developed a novel energy-based parameter that effectively correlates with experimentally observed friction coefficient patterns. Most significantly, their findings elucidated the hybrid structure's role in achieving superlubrication - specifically demonstrating how hybridization simultaneously minimizes energy fluctuations during interlayer sliding while maximizing interlayer binding energy, thereby facilitating

superlubricity. Zeolite imidazolate frameworks (ZIFs) represent a crucial subclass of metal-organic frameworks (MOFs). In their 2023 study, Li et al.[224] systematically investigated how surface frictional properties evolve in leaf-like ZIF structures through controlled functionalization and de-functionalization approaches. Their work achieved remarkable superlubricity on ZIF@mIm+eIm surfaces (where mIm = 2-methylimidazole and eIm = 2-ethylimidazole), demonstrating an ultra-low friction coefficient of 0.0076. Furthermore, the researchers elucidated a composite friction mechanism involving two key factors: (1) the anchoring effect governed by surface adhesion forces, and (2) material surface stiffness determined by coordination bond strength. Through fluorine doping, the same researchers modulated the surface tribological properties of 2D ZIFs, ultimately achieving an ultra-low friction coefficient of 0.001[225]. They demonstrated both theoretically and experimentally that fluorine doping alters electron energy dissipation pathway. Specifically, fluorination modifies the energy band structure and electrical properties of ZIFs, which simultaneously reduces energy transfer processes and suppress charge transfer by enhancing electron shielding effects. Consequently, this slows the non-radiative energy dissipation rate during energy release. In their most recent breakthrough[226], the research team successfully achieved dynamic control of 2D ZIF-8's surface tribological properties through the application of orthogonal electric fields. By employing both longitudinal and transverse field orientations, they demonstrated tunable friction coefficients spanning from 0.0037 to 0.0124. Their combined experimental and computational approach revealed that electric field regulation operates through a dual mechanism coupling interfacial adhesion with out-of-plane deformation effects. The longitudinal electric field configuration produces two concurrent effects: (1) attenuation of probe-surface anchoring forces, and (2) enhanced surface stiffness via the tight binding of interfacial charge. Meanwhile, transverse field application suppresses out-of-plane deformation through lattice stretching, providing an additional degree of control over the tribological response.

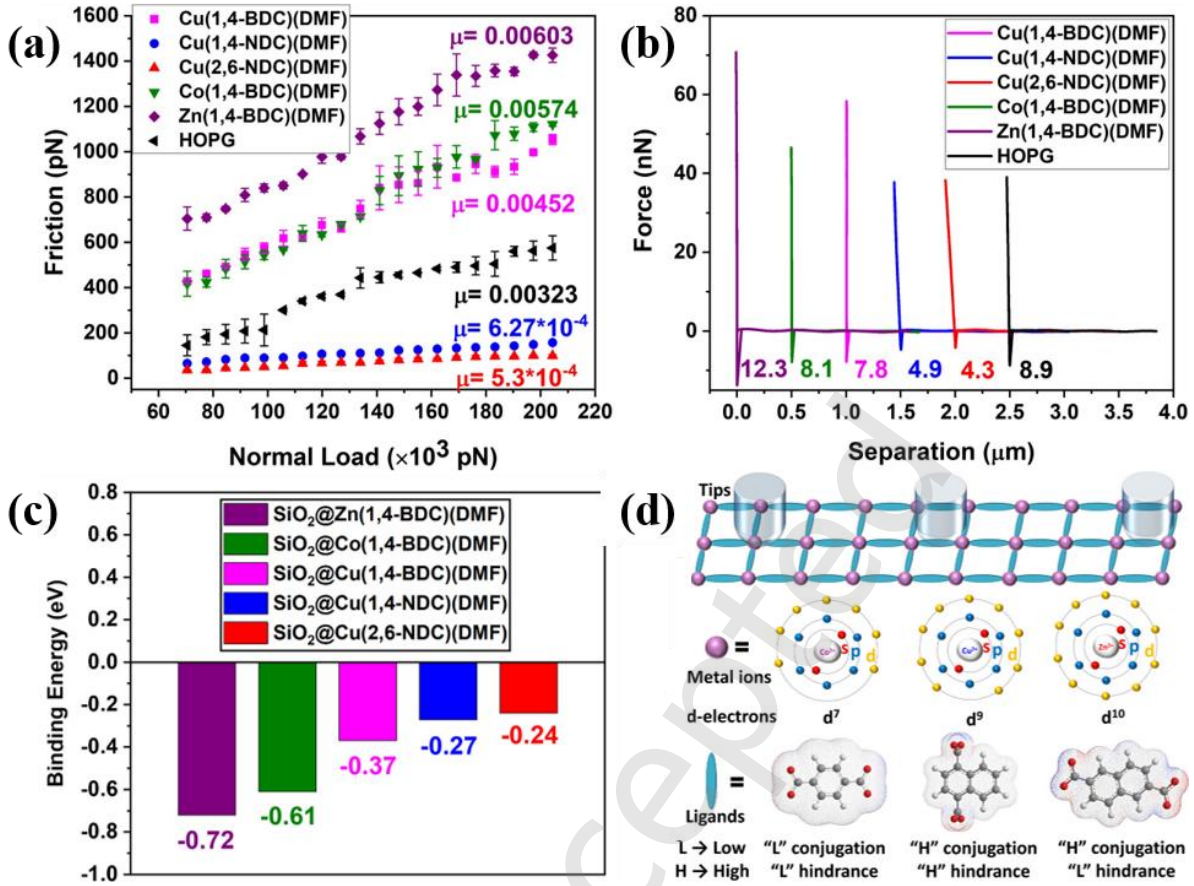

Fig. 14 (a) Frictional properties and (b) surface adhesion of isostructural MOFs with compositional variations. (c) DFT-identified anchoring mechanism for friction generation. (d) Coordination stability analysis via crystal field theory. Reprinted from ref [222] with permission from Elsevier, Copyright 2021.

The inherent lubricating properties of metal-organic frameworks (MOFs) demonstrate significant potential for applications as lubricant additives[227]. Cheng et al.[228] developed a series of two-dimensional MOFs that exhibited remarkable tribological performance when incorporated into base oils. Their research revealed that the addition of Zn(Bim)(OAc) nanosheets (where Bim represents benzimidazole and OAc denotes acetate) to liquid paraffin resulted in substantial reductions of 17.0% in friction and 22.6% in wear. Further experiments with 2D ZnBDC (BDC: 1,4-benzenedicarboxylic acid) in base oil showed even more impressive results: under ball-disc testing conditions, the average wear decreased by approximately 29%, while ball-ball testing demonstrated reductions of about 16.7% in the average friction coefficient (COF) and 20% in wear scar diameter (WSD)[229]. In 2022, Feng

et al.[230] successfully synthesized three kinds of ultrathin 2D MOF nanosheets—Ni-MOF, Zn-MOF, and Cu-MOF. These nanosheets shared identical 1,4-benzenedicarboxylate (1,4-BDC) ligands but featured different metal centers (Ni^{2+}, Zn^{2+}, and Cu^{2+}). Among them, Ni-MOF nanosheets demonstrated exceptional dispersion stability in the 500 N lubricant system, attributed to their highest absolute Zeta potential value. Tribological evaluations revealed that incorporating merely 0.1 wt% Ni-MOF nanosheets into 500 N yielded remarkable performance improvements: a 33.2 ± 12.4% reduction in friction coefficient and a 98.7 ± 0.14% decrease in wear scar diameter compared to the base lubricant. This enhancement arises from a synergistic lubrication mechanism comprising three key effects: 1) The layered structure of MOFs enables an interlayer sliding effect, facilitating smooth shear between nanosheets; 2) A polishing effect occurs as these nanosheets dynamically refine surface roughness during sliding; and 3) A unique composite tribofilm forms, where embedded iron oxide particles provide structural reinforcement, while carbonaceous and nitrogenous compounds fill interstitial gaps, resulting in a denser, smoother protective layer. In a significant advancement, Singh et al.[231] employed triethylamine, a strong electron-donating ligand, to constrain the three-dimensional growth of MOFs, successfully synthesizing ultrathin manganese-based MOF nanosheets with a mere 9-nanometer thickness. The nanostructures demonstrated exceptional tribological performance, reducing the average wear scar diameter by 35% at an optimal concentration of 0.15% w/v in paraffin oil. Surface analysis using SEM and AFM confirmed that lubricants containing 2D Mn-MOF additives substantially enhanced the surface smoothness of friction pairs. In a recent study, Zhao et al.[232] pioneered a solvent-tuned size strategy for synthesizing ultrathin Cu(2,6-NDC) nanosheets (2,6-NDC: 2,6-naphthalenedicarboxylate). These atomically thin nanostructures exhibited remarkable colloidal stability in castor oil, maintaining homogeneous dispersion without sedimentation for over 7 days. Tribological assessments conducted by a four-ball tester demonstrated that castor oil containing merely 0.08 wt% of these 2D nanosheets achieved exceptional lubrication performance. The nanofluid exhibited a 42.7% reduction in the average friction coefficient (COF) and a 58.9% decrease in wear

scar diameter (WSD) compared to pure castor oil. Surface analysis revealed a dual lubrication mechanism: (1) the low interlaminar shear strength of the 2D nanosheets facilitated smooth sliding, and (2) the abundant hydroxyl groups on the nanosheet surfaces promoted strong adsorption to metal substrates, forming a robust protective layer. This synergistic effect accounts for the dramatic improvement in tribological properties.

In 2025, Eguchi et al.[233] systematically evaluated the solid lubrication performance of 2D Cu(1,4-BDC) (1,4-BDC: benzene-1,4-dicarboxylate) using cellulose filter paper-supported specimens. Tribological tests demonstrated that Cu(1,4-BDC) exhibits outstanding lubricity with a friction coefficient (μ) of approximately 0.20, positioning it competitively between graphite (μ∼0.28) and polytetrafluoroethylene (μ∼0.14). Surface analysis of worn specimens revealed that the layered crystal structure of Cu(1,4-BDC) enables the formation of a smooth wear surface at the contact interface. In contrast, structurally analogous compounds—copper(II) benzoate and copper(II) benzene-1,3,5-tricarboxylate—exhibited significantly higher friction, underscoring the critical role of layered architecture in lubrication. Notably, thermal treatment-induced structural transformation of Cu(1,4-BDC), which introduces interlayer coordination bonds, led to an increased friction coefficient. These findings conclusively demonstrate that the exceptional solid lubricity of Cu(1,4-BDC) stems from its unique layered crystal structure, which facilitates low-friction sliding through interlayer shear and surface passivation. This study provides new insights into the design of high-performance MOF-based solid lubricants. Recently, researchers at the Vienna University of Technology successfully developed CoK-47, a novel layered titanium-based MOF, and investigated its solid lubrication capabilities[234]. Under 40% relative humidity conditions, CoK-47 demonstrated exceptional lubricating performance, achieving an ultra-low friction coefficient (μ) of approximately 0.1 - representing an 8.5-fold reduction compared to conventional lubricants. Remarkably, the material maintained stable friction coefficients between 0.1-0.2 across various material pairings, including Al_2O_3, ZrO_2, and SiC substrates. Advanced characterization techniques revealed the formation of a 240-nm amorphous tribofilm during friction,

composed primarily of titanium, oxygen, and carbon. Complementary DFT calculations suggested that water molecules actively participate in chemical reactions that facilitate CoK-47 decomposition, ultimately leading to the formation of this friction-reducing tribofilm.

7.2 Heterojunctions

Beyond the previously discussed pure materials with well-defined hybrid structures, constructing heterojunctions by assembling different types of 2D material layers has also substantially advanced superlubricity research[17]. As previously established, homojunctions exhibit periodic interlayer friction behavior governed by lattice rotational symmetry. Achieving superlubricity requires maintaining an incommensurate contact interface through controlled lattice rotation to prevent interlayer "locking". However, practical limitations emerge during sliding processes, where spontaneous lattice reorientation transitions the system into high-friction states, ultimately disrupting superlubricity[235]. Multiple factors contribute to this breakdown, including surface asperities, structural defects, and contamination[16], as well as edge effects[236] and elastic deformation-induced local commensurability[237,238]. In contrast, heterojunctions formed by dissimilar layered materials demonstrate superior resistance to crystal reorientation, enabling robust structural superlubricity and enhanced operational durability.

The intrinsic lattice mismatch between graphene and h-BN ensures incommensurability persists even when their lattice vectors are aligned. Leven et al.[239] systematically investigated how the mismatch angle influences interfacial sliding dynamics. Their research protocol involved rotating graphene to specific mismatch angles followed by lateral displacement along h-BN's chair direction, while precisely measuring the registry index amplitude along each sliding path. These measurements were subsequently plotted as characteristic curves for various graphene flake sizes at fixed mismatch angles. The study revealed two key findings: (1) the anisotropy of the sliding energy corrugation diminishes proportionally with decreasing graphene flake dimensions, and (2) optimally-sized graphene flakes exhibit near-zero sliding friction against h-BN substrates regardless of lattice

orientation, enabling robust superlubricity. In a complementary study, Mandelli et al.[240] performed comprehensive all-atom MD simulations to elucidate the fundamental mechanisms underlying the exceptional low-friction properties of graphene/h-BN heterojunctions. Their computational investigation revealed that, unlike the extensively researched twisted graphene homojunctions, the graphene/h-BN system demonstrates superior stabilization of superlubricity under high compressive loads by effectively suppressing edge atom pinning effects. The researchers identified that this remarkable friction reduction stems from the progressive formation of Moiré superlattices at the interface, which facilitates a transition from conventional viscous sliding to a soliton-like smooth motion. This phenomenon provides atomic-scale insights into the origin of ultralow friction in van der Waals heterostructures. In their groundbreaking experimental study, Ma et al.[50] quantitatively characterized the interlayer friction properties of graphite/h-BN heterostructures. The research team employed a sophisticated AFM-based measurement system where a rigid h-BN substrate was immobilized on the microscope stage, while a silica-capped graphite flake was precisely positioned atop (Figure 15a and b). Through controlled application of both normal and lateral forces via AFM tip contact with the SiO_2 coating, the researchers demonstrated two significant findings: (1) microscale single-crystal graphite/h-BN heterojunctions can maintain robust structural superlubricity even under ambient conditions and external loading, and (2) the observed friction anisotropy in these heterostructures was remarkably reduced by several orders of magnitude compared to conventional homojunction systems (Figure 15c and d). This work provides compelling experimental evidence for the exceptional tribological properties of van der Waals heterostructures.

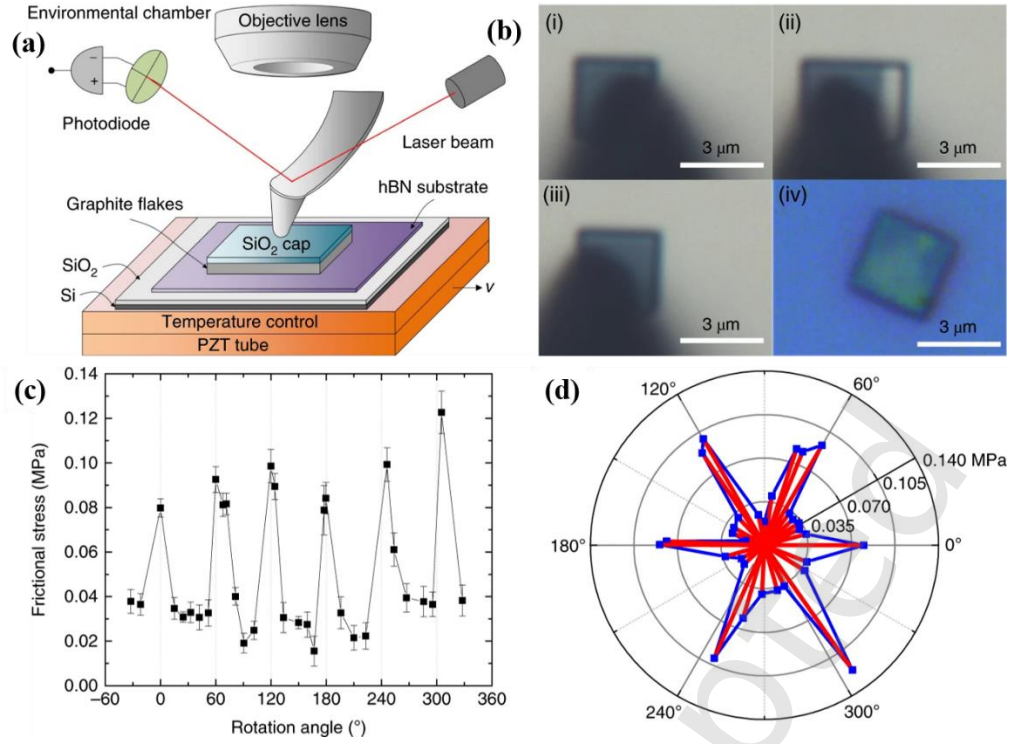

Fig. 15 (a) Schematic of the interlayer friction test device for graphite/h-BN heterojunctions, (b) Optical image of the experimental configuration, (c) Linear and (d) polar representations of frictional stress versus relative interfacial orientation. Reprinted from ref [50] with permission from Springer Nature, Copyright 2018.

The discovery of isotropic superlubricity in highly lattice-mismatched van der Waals heterostructures has opened new avenues for designing ultra-low friction interfaces. In a landmark collaboration, Zhang Guangyu's team and Tomas Polcar's group[241] employed environmental-controlled AFM to systematically investigate the frictional properties of two epitaxially grown systems: MoS_2/graphite (26.8% lattice mismatch) and MoS_2/h-BN (24.6% lattice mismatch). Their measurements revealed these heterojunctions represent ideal superlubricating systems, exhibiting both exceptionally low friction coefficients ($<10^{-6}$, approaching instrumental detection limits) and complete rotational independence (perfect isotropy). Through size-dependent studies, the researchers identified that while in-plane friction approaches zero, edge pinning effects dominate the residual friction. MD simulations elucidated the atomic-scale mechanism: lattice periodicity disruption at boundaries weakens atomic bonding, making edge atoms more mobile than their bulk counterparts. These edge

atoms exhibit both higher potential energy landscapes and greater energy dissipation during sliding, as clearly demonstrated by the computational results. This comprehensive study provides fundamental insights into the origin of friction in van der Waals heterostructures while establishing design principles for engineering superlubricating interfaces. The MoS_2/graphene van der Waals heterostructure demonstrates exceptional thermal stability in superlubricity, maintaining ultra-low friction even at elevated temperatures approaching 850 K. In their pioneering work, Yin et al.[242] performed localized thermal friction measurements revealing a counterintuitive temperature dependence - the interfacial friction coefficient exhibited further reduction with increasing temperature. Their comprehensive analysis identified MoS_2 edge effects as the primary contributor to residual friction. The proposed multi-contact model attributes this anomalous behavior to thermally-activated breaking of edge contacts, which facilitates interfacial sliding. Remarkably, the heterostructure showed outstanding durability, with over 1000 high-temperature friction cycles producing negligible mechanical degradation or chemical modification, highlighting its potential for extreme-condition applications.

Researchers evaluated the tribological properties of MXene heterojunction systems at the atomic scale using DFT and found that the Ti_2CO_2/MoS_2 heterojunction interface exhibits an extremely low sliding activation energy barrier of approximately 0.015 eV per oxygen atom, achieving a superlubricity state[243]. This behavior stems from the heterojunction interface disrupting interlayer charge symmetry (weakening charge interlocking) and promoting interface charge redistribution, thereby reducing the sliding activation energy barrier. Due to this charge redistribution, the intermediate layer becomes more prone to sliding when normal force decreases, resulting in ultra-low friction. In this state, interlayer charge interlocking and charge redistribution compete, jointly determining the magnitude of the sliding activation energy barrier and causing irregular fluctuations in the friction coefficient. Ideal superlubricity behavior can be achieved by optimizing interfacial charge concentration while preventing charge transfer to sub-surface layers. Mochalin et al.[103]

experimentally verified the superlubricity capability of MXene-based 2D heterojunction interfaces through atomic-scale friction measurements. Their results showed that MXene/graphene and MXene/$MoSe_2$ heterostructures achieved remarkably low friction coefficients ($\mu < 0.01$) under a normal load of ~0.91 μN across a 15 nm sliding distance. The study further revealed that increasing the number of stacked layers enhances the out-of-plane stiffness, which significantly mitigates interfacial elastic deformation. This stiffness effect plays a pivotal role in suppressing the characteristic "wrinkling effect" of 2D materials, consequently maintaining stable ultra-low friction conditions.

In a recent study, Xu et al.[244] employed molecular dynamics simulations to investigate atomic-scale tribological behavior in black phosphorus/graphene heterostructures, examining the effects of rotation angle, temperature, and sliding velocity. Their results demonstrate that the transition from commensurate to incommensurate contact states reduces the friction coefficient by approximately an order of magnitude. The heterostructures also exhibit improved stress distribution, as evidenced by Von Mises stress analysis. Furthermore, observed changes in energy barriers and potential energy surfaces show positive correlation with corresponding variations in the friction coefficient.

Li et al.[245] developed a novel strategy to scale up superlubricity from microscopic contact points to macroscopic engineering applications. Their approach involved engineering a micro-convex surface topography on steel substrates, effectively transforming macroscopic surface contacts into numerous microscopic point contacts. At each contact point, they implemented a combination of precision pre-grinding and deposition of 2D heterojunction coatings, including both covalent/ionic (graphene/α-ZrP) and covalent/covalent (graphene/MoS_2, h-BN/MoS_2, MoS_2/WS_2) structural combinations. This interfacial engineering achieved three critical superlubricity prerequisites: (1) ordered lamellar stacking, (2) weakened chemical interactions, and (3) incommensurable contact configurations. The resulting steel-steel tribosystem demonstrated remarkable macroscopic superlubricity performance, featuring: an ultra-low friction coefficient ($\mu \sim 10^{-3}$), exceptional durability ($>1\times10^6$ cycles), and broad

material/load adaptability. This breakthrough represents a significant advancement toward industrial-scale superlubrication applications.

8 Conclusion and Perspectives

This review comprehensively summarizes significant advances in tribological research on 2D materials across various structural classifications, including inorganic, organic, carbon-based, and hybrid systems. The discussion encompasses both fundamental studies (spanning atomic-scale friction mechanisms and superlubricity phenomena) and applied research (covering lubricant additives, solid lubricating coatings, and composite reinforcement applications). While significant progress has been achieved in 2D material tribology, critical knowledge gaps and technical challenges remain that limit both fundamental mechanistic understanding and practical large-scale implementations.

1. Despite the remarkable structural diversity reported in current literature, the fundamental structure-performance relationships remain inadequately understood. While Liu et al.[47] have made pioneering contributions by establishing correlations between structural factors and interlayer friction mechanisms, several critical scientific questions persist. Notably, the precise response mechanisms linking interlayer friction resistance and adhesion forces with atomic-scale structural details require further elucidation. A comprehensive understanding demands systematic approaches involving: (1) precise structural variable design, (2) quantitative mapping of structure-tribological property correlations, and (3) first-principles computational analysis of how structural parameters govern key indicators like sliding energy barriers. Only through such multidimensional investigations can we achieve accurate structure-performance relationship models.

2. Recent advances in 2D lubricating materials have highlighted the exceptional potential of organic and inorganic-organic hybrid structures as additives or reinforcement phases. Their molecular-level structural compatibility with lubricants/polymer matrices, coupled with abundant chemically modifiable sites, offers inherent advantages for enhancing matrix affinity and dispersion stability. Nevertheless, significant research gaps persist in this emerging field. (1) For inorganic-organic hybrid

structures like MOFs, while preliminary understanding of their microscopic friction mechanisms exists, fundamental investigations under practical macroscopic conditions remain scarce. Crucially, macroscopic tribological performance depends not merely on intrinsic lubricity ability but also on system-level synergies - particularly tribofilm formation and transfer dynamics. Promisingly, the demonstrated catalytic activity of MOFs[246-249] suggests potential for catalytic generation of tribofilm. (2) Regarding COF-based additives, despite extensive studies of their tribological properties, no consensus exists on optimal structural selection criteria. This stems from insufficient fundamental understanding of their atomic-scale friction mechanisms and intrinsic structure-performance relationships. Addressing these knowledge gaps would substantially advance the systematic development of 2D materials for tribological applications.

3. While conventional 2D materials like graphene and molybdenum disulfide benefit from “top-down” synthesis approaches using mineral precursors that facilitate scalable production[250-253], emerging 2D lubricating structures face more complex fabrication challenges. The synthesis of advanced frameworks such as COFs and MOFs typically requires spatial confinement strategies[37,254], creating a fundamental trade-off between yield, quality, and production costs - often referred to as the "impossible triangle" in materials manufacturing. Overcoming this limitation through the development of cost-effective, large-scale synthesis methods remains crucial for universal implementation. Notably, the higher production costs of these novel lubricants may be justified by targeting specialized applications where conventional materials underperform. In high-end mechanical systems, for instance, functional interfaces increasingly demand multifunctional lubrication solutions. A representative case is high-speed train pantographs, where coatings must simultaneously achieve exceptional wear resistance, efficient lubrication, and high electrical conductivity. The highest electrical conductivity reported for MOFs to date is 1580 $S \cdot m^{-1}$[255], which is comparable to that of graphite and potentially usable as a functional coating for the pantographs. Investigating the stimulus-responsive behavior of these advanced lubricants and elucidating the coupling mechanisms between

external stimuli and intrinsic tribological properties could enable the rational design of next-generation smart lubrication systems.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Lei Liu was recruited as a Class A talent to join the School of Mechanical Engineering at North China University of Water Resources and Electric Power in 2025. He obtained his doctoral degree from the State Key Laboratory of Tribology in Advanced Equipment at Tsinghua University and subsequently conducted postdoctoral research at the institution. His research focuses on the innovative design of lubricant additive for high-end equipment. His current H-index stands at 20.

Linjian Shangguan serves as the leader of the Henan Higher Education Institutions' Scientific Innovation Team for 'Engineering Machinery High Reliability Design and Assurance'. He also holds the position of director at the Henan Research Center for Smart Lubrication and Equipment Health Management Technology. With extensive experience in his field, Professor Shangguan has dedicated his career to researching large-scale construction equipment structural design and developing technologies to ensure equipment operational reliability.

Han Peng is a professor at the School of Mechanical Engineering, North China University of Water Resources and Electric Power. Based at the Henan Provincial Engineering Technology Research Center for Smart Lubrication and Equipment Health Management and the Zhengzhou Key Laboratory of Construction Machinery Lubrication Technology, she specializes in research and teaching in fields including: Energy-efficient lubrication analysis and design for mechanical vehicles; Intelligent operation analysis and control of machinery; Equipment wear mechanism analysis and intelligent lubrication technologies.

Yuhong Liu currently holds the position of Professor at Tsinghua University's State Key Laboratory of Tribology in Advanced Equipment. In recognition of her outstanding contributions, she was honored with the NSFC Distinguished Young Scholar Fund in 2016. Her ongoing research focuses on superlubricity and drag reduction.