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Review

Vanishing Friction: Progress toward Mechanistic Understanding and Potential Engineering Applications

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Abstract

Friction and wear collectively account for nearly a quarter of the world's energy consumption, resulting in over eight Gigatons of CO_2 emissions annually. With increasing mobility and industrial activity, the adverse effects of friction and wear on energy, the environment, the global economy, and sustainability will undoubtedly intensify. Unless we reverse this unsustainable trend, our planet could face a major ecological and environmental catastrophe. Fortunately, significant strides have been made in reducing friction to almost undetectable levels, with friction coefficients below 0.001. These remarkable achievements have resulted from numerous collaborative efforts and global initiatives focused on developing novel materials, surfaces, and interfaces that exhibit minimal or near zero friction, even at macro or engineering scales. This paper provides a comprehensive overview of the factors that contribute to and hinder superlubric sliding conditions, examining the impact of both intrinsic and extrinsic factors that are integral parts of the test conditions and environments. Drawing from recent analytical, experimental, and computational findings, underlying mechanisms most responsible for superlubricity are also discussed. The paper discusses recent mechanistic studies on highly ordered 2D materials, such as graphene, MoS_2, h-BN, MXene, etc., and thin solid coatings such as diamond-like carbon or DLCs, as well as liquids, and discusses their potential for the development of large-scale mechanical systems. These exciting advancements pave the way for designing and producing next-generation engineering systems that can minimize friction in practical applications, thus conserving energy, enhancing durability, and protecting the environment for a sustainable future.

Keywords

superlubricity, 2D materials, friction, MXene, diamond-like carbon

1 Introduction

Superlubricity in tribology, often referred to as a state in which friction or resistance to sliding between two solid surfaces essentially disappears, has been a long-standing aspiration for tribologists not only because of its profound scientific significance but also its enormous industrial implications. Specifically, if this state could be achieved practically across all moving mechanical assemblies worldwide, it would undeniably have a significant positive impact on our global energy and environmental sustainability goals, as energy losses due to friction and wear in such assemblies are estimated to contribute to nearly a quarter of the world's total energy output and account for more than eight Gigatons of CO_2 emissions annually [1]. Further reducing friction and wear is imperative for achieving much greater efficiency and reliability in future mechanical systems. Therefore, the research on superlubricity is critically important and has been gaining momentum over the years (Fig. 1).

Since the early 1990s, scientists and engineers have been exploring ways to eliminate sources of friction and thus achieve superlubricity. Initially, there was relatively low scientific and industrial interest, with much of the research focused on understanding the atomistic origins of superlubricity. Reflecting on the substantial growth in superlubricity research, thousands of publications have since emerged on this topic in reputable journals, conference proceedings, review articles, and dedicated books [2-11]. With this increasing interest and progress, there is hope that we may witness the operation of moving mechanical assemblies with minimal friction, thus consuming significantly less energy and producing fewer harmful emissions.

In several natural or artificial tribological systems, superlubricity or frictionless sliding already exists. For example, in articulated joints, journal or foil bearings, and magnetically levitated surfaces, the frictional energy dissipation is minimal and mainly restricted to the shearing of the fluid media due to

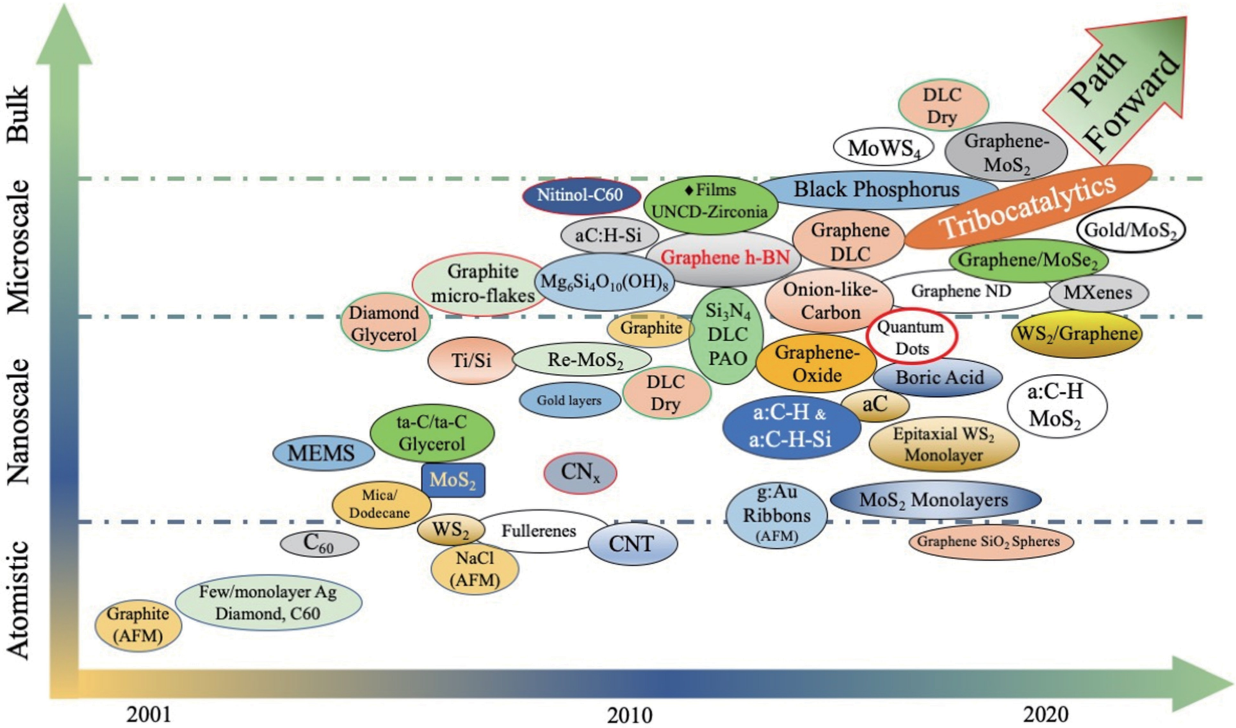

Fig. 1 Chronological evolution of superlubricity over length scales. Reproduced with permission from [2] Copyright (2022) Frontiers.

their viscosity. Nanoscale levitation, driven by Casimir forces, may also create ideal conditions for achieving superlubricity or near-frictionless sliding.

Thanks to significant advances in computational modeling and simulation methods in recent years, we are better equipped to understand the atomistic origins of superlubricity and use this knowledge to design tribological systems that can offer superlubricity across a wider range of environments and tribological conditions. Incommensurability between 2D materials has long been predicted, simulated, and experimentally verified as a major superlubricity mechanism in graphite, graphene, MoS_2, and other 2D materials. Researchers have independently demonstrated that friction vanishes by precisely controlling the twisting or rotating of 2D materials' atomic planes from complete commensurability to complete incommensurability [12-14]. With total incommensurability, where superlubricity comes to life, surface atoms are arranged in a way that prevents direct registry or contact, which diminishes interatomic interactions and, hence, adhesion and friction. While these scientific discoveries are indeed exciting in the field of tribology, significant challenges still exist in achieving superlubricity at various scales in real or industrial systems. For example, obtaining and maintaining an extremely smooth surface and continuous incommensurability in real tribological systems poses significant difficulty. Gaseous, liquid, or solid contaminations are also very hard to avoid as they can easily interfere with the very delicate nature of superlubricity.

In the following sections, we present a comprehensive and up-to-date review of recent exciting developments in superlubricity. Special emphasis is placed on the most important mechanisms and sliding conditions that govern superlubricity, how intrinsic and extrinsic factors affect adhesion and friction between solid surfaces, and how they can be minimized to achieve superlubricity. As special case studies, we delve into the recent mechanistic understanding of the superlubricity in traditional and emerging 2D materials such as graphene, MoS_2, h-BN, MXene, black phosphorous, etc., as well as thin solid coatings, such as DLC coatings and sliding systems involving liquids. We also assess the possibility of achieving macro-scale superlubricity in large-scale mechanical systems.

2 A brief historical perspective

2.1 Early studies

The word "superlubricity" was first coined by the late Prof. Motohisa Hirano in the early 1990s and the key underlying mechanisms were described by his research team in references [15-17]. Using mica as an atomically smooth and defect-free substrate, they confirmed the existence of superlubricity through nanoscale friction tests, where they showed that friction can vanish by adjusting the degree of lattice misfit or incommensurability. Motivated by Hirano's original work, Martin et al., further confirmed the existence of superlubricity in MoS_2 using an ultra-high-vacuum tribometer under conditions where incommensurability was achieved [18-20]. During the remainder of the 1990s, further progress was made both in the theoretical understanding and experimental verification of superlubricity by these and several other authors [21-23]. In fact, dedicated experimental research has shown that in addition to incommensurability, the specific chemistry or chemical composition of the test environment may also play a significant role in the superlubricity of MoS_2 [20].

2.2 Diamond-like carbon coatings

Superlow friction of diamond-like carbon films was reported in the 1990s, although these films were structurally disordered or amorphous, so the incommensurability mechanism would not apply. However, during the second half of the 1990s, DLC films with friction coefficients below 0.01 were developed. Specifically, it was found that the superlubric sliding behaviors of these films were very dependent on the chemistry of gas discharge plasmas from which they are extracted and the test environments or conditions under which they were tested. In general, superlow friction, or superlubricity, was only achieved in inert gas environments with highly hydrogenated DLCs, while hydrogen-free or non-hydrogenated DLCs' friction coefficients were among the highest in such inert test environments [24-34]. Figure 2 shows the typical superlubric behavior of a highly hydrogenated DLC, along with its proposed lubrication mechanism.

In other studies, it was further confirmed that the high hydrogen content of the DLCs or the test environment was critically important for attaining superlubricity [35-39]. Recent studies have claimed that highly hydrogenated DLC films can provide friction coefficients even lower than 0.001 when sliding against zirconia balls [40, 41]. Such a frictional behavior was thought to result from the formation of a polymer-like tribofilm, especially under extreme contact pressures (i.e., 2.6 GPa), and by a unique catalytic effect afforded by counterface zirconia. Throughout the 2000s, the fundamental mechanisms of superlubricity of DLCs and other materials have been further explored using more advanced computational, experimental, and surface analytical tools. As a result, further insights were gained into what contributes to and compromises superlubricity in such carbon films [42-47].

Much of the computational efforts on the superlubric sliding behaviors of DLC coatings were directed toward the mechanistic understanding of their sliding behaviors in different environments. Earlier studies by Dag et al. [48] have confirmed diamond and DLC films' surface termination states can play a major role in their frictional behaviors. Consistent with the proposed lubrication mechanism in Fig. 2b., they showed that when hydrogen atoms terminate the sliding surfaces of such films, the surface becomes highly positively charged and, due to the creation of a dipole configuration at the sliding contact interface, such positively charged surfaces can then lead to the generation of repulsive forces at the contact interface, thus reducing adhesion and, hence, friction. Other researchers have also reached similar conclusions regarding the effects of hydrogen termination of diamond and DLC films [49-52]. Specifically, they have shown that hydrogen termination was critically important for achieving and maintaining low friction and wear on sliding DLC and diamond surfaces.

After having their superlubricity well-demonstrated throughout the 2000s, DLC films became the focal points of all kinds of industrial applications, including engines, orthopedics, optics, and various other applications [53-64]. Since these original studies, thousands of new research papers have been produced on the tribological properties and diverse applications of DLC films. Thanks to these studies, DLC films are used today in a broad spectrum of industrial applications with great success and excellent protection against environmental and mechanical degradations [65-71].

In addition to highly hydrogenated DLCs, some doped DLCs were shown to provide superlubricity. Some of these include carbon nitride (CNx) coatings, which were shown to attain superlubricity after a brief run-in or surface conditioning period in oxidizing environments. During the conditioning period friction could be very high, but after switching to a dry nitrogen environment, friction coefficients as low as 0.005 were shown to be feasible [72]. Such a dramatic reduction in friction has been attributed to the formation of an extremely shearable tribolayer accommodating sliding velocity without creating much friction. In other DLCs doped with silicon, superlow friction was also achieved when tested under high vacuum conditions [73, 74]. Their superlubricity mechanisms are not yet well understood but as in CNx, they were attributed to the formation of a highly shearable transfer layer. Fluorine doping of DLC was also shown to provide very low friction coefficients. Sliding tests on such DLCs could provide friction coefficients down to 0.005 in ultra-high vacuum [75]. Other DLCs where superlubricity was observed include fullerene-like hydrogen-free and hydrogenated DLC films [76], as well as sulfur-doped DLCs [77].

Despite more than three decades since the first reporting of their superlubricity, DLC films continue to enjoy strong interest from both the research and industrial communities for their fascinating ultra-low friction and wear behaviors. Numerous research and review articles have dealt with their superlubricity, including new insights into their lubrication mechanisms [78-82].

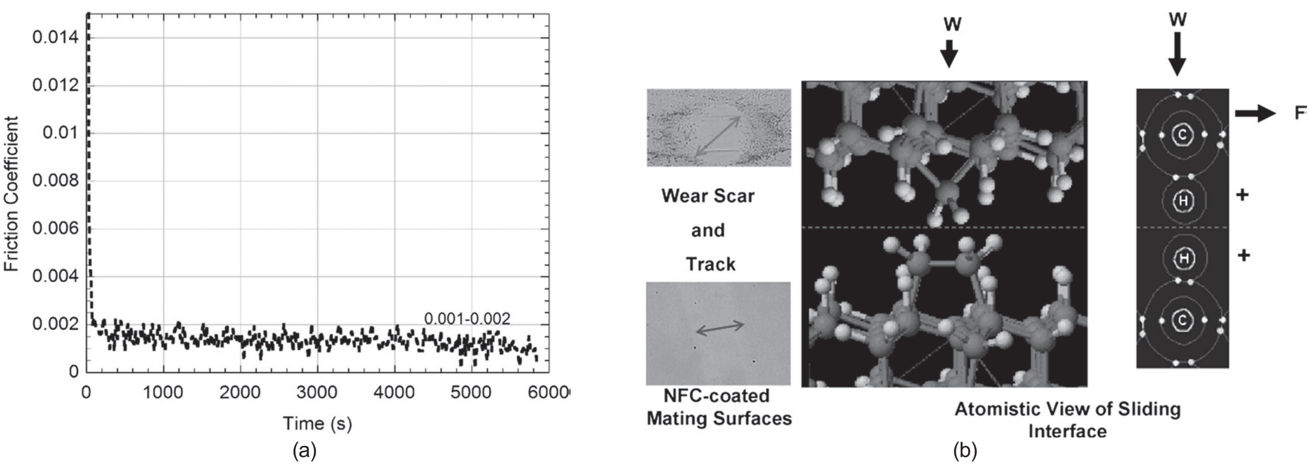

Fig. 2 Superlow friction of a highly hydrogenated DLC film (a) and a mechanistic illustration of its superlow friction behavior. Adapted with permission from [27] Copyright (2000) American Institute of Physics and [30] Copyright (2001) Elsevier.

2.3 2D materials

Work on the superlubricity of bulk lamellar or 2D materials goes back to the 1990s. The earliest reporting of superlubric sliding behavior in such materials is attributed to natural graphite. This credit goes to Prof. David Tabor and his team, who demonstrated that the basal planes of natural graphite are very slick, yielding friction coefficients in the range of 0.005 to 0.02 under light loads [83]. Using an AFM instrument, Mate et al. have also shown that friction coefficients of 0.005 to 0.015 are feasible against a tungsten tip when slid against the basal planes of a graphite surface at low loads [84]. As mentioned earlier, another well-known lamellar solid, MoS_2, was also shown to exhibit superlubricity by Martin et al. under incommensurability conditions in high vacuum. More systematic studies by Dienwiebel et al. [14, 85] showed that just like MoS_2, the friction of graphite surfaces depends very closely on structural orientation at atomic scales [86]. Specifically, its friction coefficient changes dramatically with the rotation angle between the top and bottom graphite surfaces. Friction is relatively high at 0 and 60° rotation angles, but in between, friction goes down literally to zero, suggesting that mechanistically, the superlubricity of graphite largely results from a state of incommensurate contact between sliding graphite surfaces.

Intercalation of graphite layers with C60 was also shown to provide superlubricity. Specifically, theoretical and nanoscale experimental studies by Miura et al. confirmed that it is possible to achieve a nearly frictionless sliding regime with graphite sheets if C60 is used as intercalates between their alternating layers [87-89]. C60 was thought to increase the interlayer spacing between graphite layers and thus further diminish interlayer bonding while acting as a molecular-scale ball bearing.

Other researchers have also demonstrated the pivotal role of incommensurability in the superlubricity of more exotic 2D materials. With the discovery of graphene and other 2D materials, the incommensurability mechanism of superlubricity was further confirmed by both computational and dedicated experimental studies [90-93]. Besides the structural origins of superlubricity of 2D materials in nano to micro-scale experiments, researchers have shown that graphene can also achieve macro-scale superlubricity when combined with highly hydrogenated DLC and diamond nanoparticles [94]. Computer simulations and structural studies have shown that the formation of nanoscrolls achieved such a superlubric sliding regime by simply wrapping graphene sheets around nanodiamond particles of 2 to 4 nm in size. These scrolls were then able to separate the sliding surfaces and roll like nanoscale ball bearings to accommodate sliding motion and thus decrease the friction coefficient down to 0.004. Likewise, by replacing graphene with MoS_2 flakes or nanodiamonds with iron nanoparticles within the same test system, superlow friction values were achieved through special tribochemical processes that converted initial carbon form into onion-like carbon structures, which were also very capable of producing similar graphene+diamond nanoscroll effects and thus achieving superlubricity [95, 96]. Successful demonstration of superlubricity with 2D materials has been further demonstrated for multiple systems enabling control of the incommensurability regime (Fig. 3).

Recent tribological research on emerging 2D materials like MXenes, black phosphorous, and other metal dichalcogenides has confirmed superlubricity when mixed or further

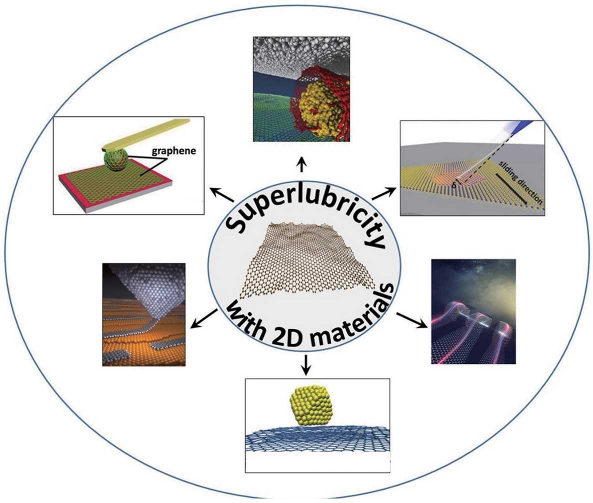

Fig. 3 Examples of observed superlubricity received with 2D materials in different systems. Adapted with permission from [86] Copyright (2018) American Institute of Physics.

functionalized with certain polar fluids in a nano-colloidal dispersion [97-99]. It seems that when such polar molecules (such as ionic liquids, various glycols, aqueous acid-based solutions, etc.) are used on certain 2D materials as additives in liquid lubricating media, a significantly higher or beneficial hydrodynamic effect is achieved through a hydration mechanism leading to liquid superlubricity [100-104]. The presence or availability of hydrogen ions is extremely important for achieving superlubricity in certain sliding systems as hydrogen in an ionized state has its positively charged core sticking out to the surface by creating a positively charged double layer, hence the hydration effect leading to ultralow friction [105-108]. Superlubricity in other liquid media, including alcohols, glycols, glycerols, ionic liquids, hydrogels, etc., was also achieved using a wide range of material pairs at macro-scales and attributed to the extent and chemical nature of tribofilms resulting from such media [109-113].

2.4 Other materials systems affording superlubricity

A host of heterostructures in combination with 2D materials were also shown to provide superlubricity under dry and lubricated conditions [114-121]. In most of these systems, superlubricity was attributed to the existence of a very favorable or enhanced state of incommensurability due to lattice misfit. The degree and distribution of such misfits were claimed to play a dominant role in the extent of superlubricity being provided by such heterostructures. These results indicate that in actual engineering applications, using or pairing two distinct or different 2D materials at the sliding contact interfaces will be highly desirable. Here, special attention should be given to the systems demonstrating superlubricity under elevated temperature conditions due to self-adaptation [122] or tribocatalysis [123] processes. Superlubricity has been observed in the presence of magnesium silicate hydrate (MSH) mixed with antimony oxide and MoS_2 nanopowders and burnished on nickel superalloy or copper substrates when testing at temperatures above 200°C [124, 125].

Black phosphorous, also known as phosphorene, combines a host of unusual physical, electrochemical, electrical, and optical properties, making it attractive for various industrial applications, including batteries, sensors, transistors, photovoltaics, and other applications. In particular, high electrical conductivity makes it an ideal candidate for electrical applications. The ease of chemical functionalization combined with a 2D structure of black phosphorous opened up the possibility of using it as a low-friction tribomaterial [126]. As with other nanomaterials, black phosphorous sheets can be functionalized and used as a colloidal lubricant additive. A systematic study compared the tribological performance of graphene oxide and MoS_2 nanosheets against the black phosphorous nanosheets. Even at the lowest concentration (i.e., 0.1 ppm), favorable lubricity was achieved with black phosphorous [127]. When mixed with oleic acid, black phosphorus reduced friction from about 0.1 to 0.006 at a concentration of 0.1 mass% [128]. Recent studies have shown that the use of partially oxidized black phosphorus in an oleic acid-containing oil may favor the creation of a special tribofilm consisting of amorphous carbon, BP crystal, and phosphorus oxide, which together enables macroscale superlubricity [129].

MXenes are another class of novel 2D materials based on transition-metal carbides, nitrides, or carbonitrides built up based on MAX-phases [130]. Their potential for chemical, electrochemical, and electrical applications has been well-recognized and exploited in recent years [131]. Their 2D architectures can also make them a prime candidate for tribological applications. The friction and wear performance of fine powders of MXene were either comparable or slightly better than graphite when tested under the same conditions [132]. In other studies, the friction-induced graphitization of Ti_3C_2-based MXene led to much-reduced friction and wear [133]. These and other recent studies have confirmed that MXene alone or combined with other lubricious materials offer great potential as a solid lubricant [134-136]. However, achieving superlubricity with MXenes required using DLC, MoS_2, or polar molecules like glycerol. When tested against DLC, Ti_3C_2 MXene deposited on Si substrates could afford friction coefficients down to 0.006 in dry nitrogen [137]. Likewise, when the same type of MXene was mixed with glycerol, friction coefficients of 0.002 were achieved [138]. In both cases, the roughness of the substrate materials was very low, ensuring precise control of the sliding interfaces. This challenge has been overcome recently by mixing MXene with MoS_2 and spray-coating the mixture on rough steel surfaces (Fig. 4). During sliding against the steel counter body under high load and sliding speed conditions, the coating transformed into the robust tribofilm with reoriented MoS_2 and MXene basal planes, creating local incommensurability states leading to vanishing friction and wear [139].

3 Summary and prospects for real engineering applications

Superlubricity remains an area of active research and development requiring innovative materials and interdisciplinary collaboration to overcome the existing barriers to its practical implementation across various industrial and technological sectors. In a world where energy conservation is paramount, minimizing friction losses could lead to substantial energy savings, lower operational costs, and reduced carbon emissions. Minimized friction means less mechanical stress and often less wear, translating to longer equipment lifespans and decreased maintenance requirements. At the same time, superlubricity opens new opportunities for designing high-performance systems that were previously unattainable due to friction-related limitations.

Through intensified research efforts in recent years, significant progress has been made in gaining a fundamental understanding of superlubricity and translating this knowledge for its experimental demonstration. The discovery of graphene and other 2D materials, with their atomically smooth surfaces, created an ideal platform for precise design and nano to macro-scale demonstrations of superlubricity in various systems, configurations, and operating conditions.

However, while these scientific outputs and the prospects of implementing superlubricity are highly promising, several challenges remain. Achieving superlubricity at the laboratory level is one thing, but scaling up the technology to produce millions of mechanical assemblies is another. Consistency of the performance and durability over the lifespan of the assembly, which can reach millions of hours of operation or several hundred thousand miles, is a major concern.

The successful research examples are based on highly idealized smooth, clean, and defect-free surfaces; however, most real surfaces in industrial machinery are rarely perfect. Moreover, contamination or wear debris can easily disrupt superlubricity. At the same time, the variety of practical temperature and humidity settings creates another challenge and requires adapting the superlubricity to sustain transitions between a broader range of environmental conditions.

New sustainable transportation needs, such as electric vehicles (EVs), create additional exciting challenges for tribologists. As the world rapidly transitions from internal combustion engines to electric propulsion, realizing superlubricity in such systems could lead to significant reductions in friction within electric motors and drivetrains. This achievement can undoubtedly result in lower energy consumption, extending the vehicle's range on a single charge and making electric vehicles (EVs) the greenest and most appealing form of transportation for consumers. However, in the case of EVs, the lubricants and mechanical stresses often experience electrified conditions and high-temperature variations while in operation. Therefore, the existing knowledge about superlubricity requires further advancements to accommodate such electrified environments and the harsher operating conditions of such EVs.

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Financial interest statement

The authors declare no competing financial interest.

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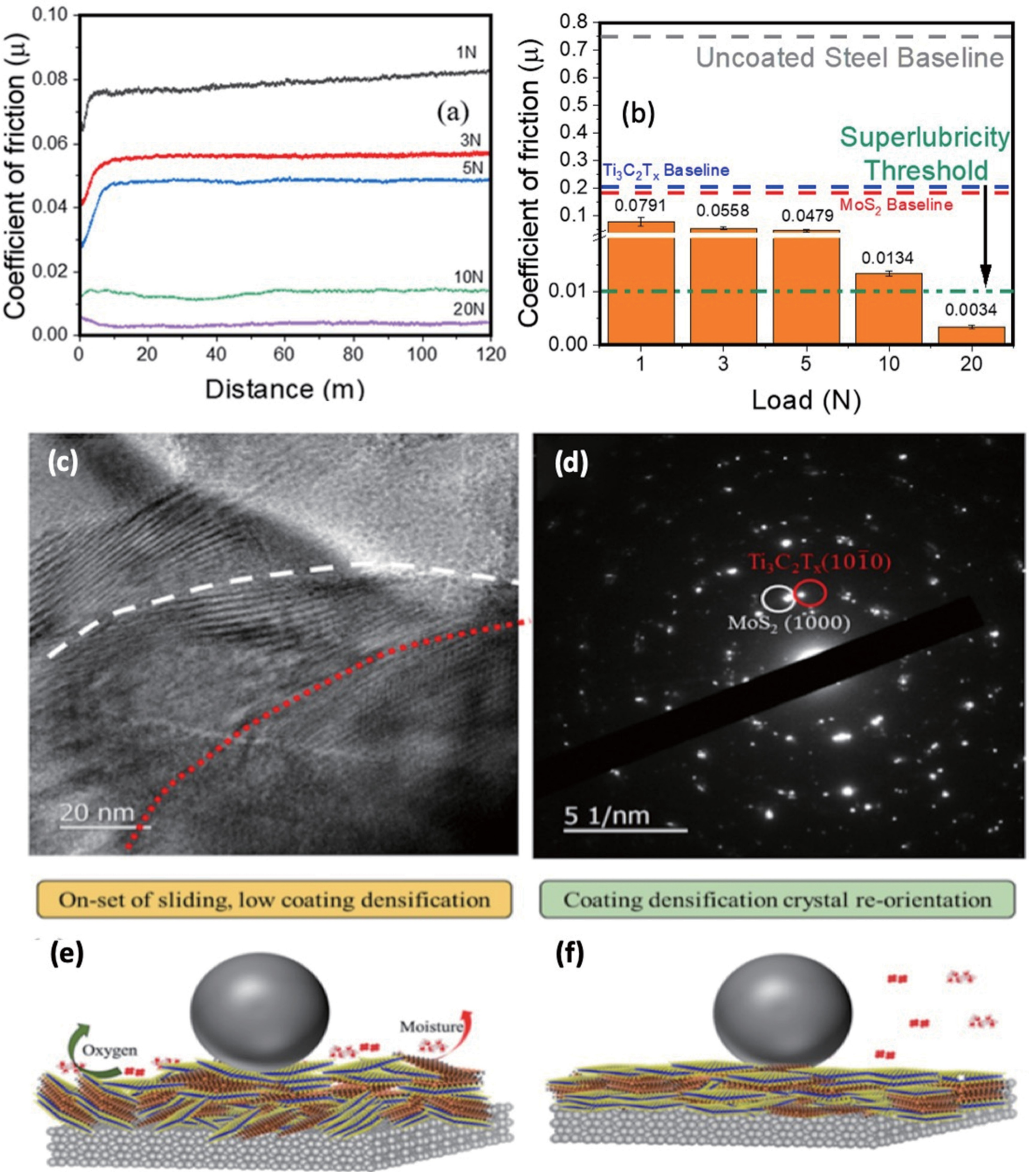

Fig. 4 Superlubricity with MXene. (a) Coefficient of friction behavior showing the performance of MoS_2-Ti_3C_2Tx solid lubricant coatings under unidirectional sliding at 0.1 m/s under various contact loads as a function of sliding distance. (b) Summary of steady-state friction values juxtaposed with steel-on-steel reference. Bright field TEM images of tribolayer at (c) at the end of the long-term test and (d) electron diffraction image. Schematic of an in-operando mechanism consisting of coating densification and reorientation from (e) the original mixture to (f) the state that resulted in superlubricity. Adapted with permission from [139] Copyright (2023) American Chemical Society.

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