

The art of friction control: Advanced diamond-like carbon coating design for aerospace tribology

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Abstract: In the aerospace sector, where extreme service conditions severely challenge the reliability and longevity of critical components, diamond-like carbon (DLC) coatings offer significant promise due to their superior tribological properties, chemical stability, and mechanical strength. However, their broader application is hindered by intrinsic limitations such as high residual stress, low fracture toughness, and strong environmental sensitivity of tribological behavior, which collectively restrict their high-performance deployment in aerospace systems. This review offers a thorough examination of recent advancements in the tribological behavior of DLC coatings under extreme environmental conditions. It begins with an examination of the coatings' sensitivity to temperature, atmospheric composition, and humidity, along with the mechanistic principles underlying these environmental

effects. Next, pivotal strategies for enhancing tribological performance are examined, encompassing layered structural design, compositional tuning, surface functionalization, and solid-liquid lubrication regulation, with an emphasis on their physicochemical mechanisms. The current status and challenges of DLC coating applications in critical aerospace components are then assessed. In response to the mounting demand for high reliability and multifunctionality in extreme aerospace environments, the review outlines several prospective research directions: optimization of micro/nano-scale architectures and interfaces, development of self-healing DLC functional interfaces, construction of multifunctional composite DLC systems, and implementation of intelligent, environmentally adaptive surface engineering strategies. Together, these endeavors aspire to establish a theoretical and technological foundation for the reliable, long-term service of aerospace systems under harsh conditions.

Keywords: DLC coating, aerospace sector, tribological performance, environmental sensitivity, performance regulation

1 Introduction

In the realm of aerospace engineering, friction, wear, and lubrication failure represent fundamental challenges that critically dictate the performance, reliability, and longevity of vital component [1-5], particularly under extreme aerospace environments. These environments mainly include low-Earth orbit, deep-space exploration, and atmospheric re-entry scenarios, each characterized by distinct combinations of temperature, pressure, radiation, and gaseous conditions [6, 7]. For instance, low-Earth orbit environments involve alternating thermal cycling and atomic oxygen exposure, deep-space environments are dominated by ultralow temperatures and high vacuum conditions, whereas atmospheric re-entry subjects components to intense aerodynamic heating and oxidative atmospheres. As critical components are routinely subjected to such harsh conditions over prolonged periods, their lubrication performance is profoundly compromised, resulting in a marked increase in friction

coefficients and consequently accelerating wear [8, 9]. The friction, wear, and lubrication failure issues under these extreme conditions impose significant constraints on the operational stability and service life of aerospace systems [10-14]. In practical aerospace applications, coating failure may manifest through different forms, including excessive friction increase, severe wear, coating delamination, substrate exposure, or the loss of intended tribological functions, depending on specific service requirements and operating conditions. Therefore, developing advanced lubrication materials and technologies capable of adapting to extreme service environments has become an urgent demand in aerospace engineering [6, 12, 15-18].

Compared with traditional liquid lubricants, solid lubricant materials exhibit superior environmental adaptability and outstanding stability under extreme operating conditions [19-25]. Among them, diamond-like carbon (DLC) coatings have attracted extensive attention because of their unique combination of ultra-high hardness, low friction coefficient, excellent wear resistance, and chemical inertness [26-29]. DLC coatings are amorphous carbon-based materials composed of mixed sp^2 and sp^3 hybridized carbon structures, which provide both high mechanical strength and low shear resistance [30-37]. As illustrated in Fig. 1, owing to these advantages, DLC coatings have been widely investigated and applied in various fields, particularly in aerospace, automotive, precision manufacturing, biomedical engineering, and energy industries, where reliable friction reduction and wear resistance are critically required under harsh service conditions [38, 39].

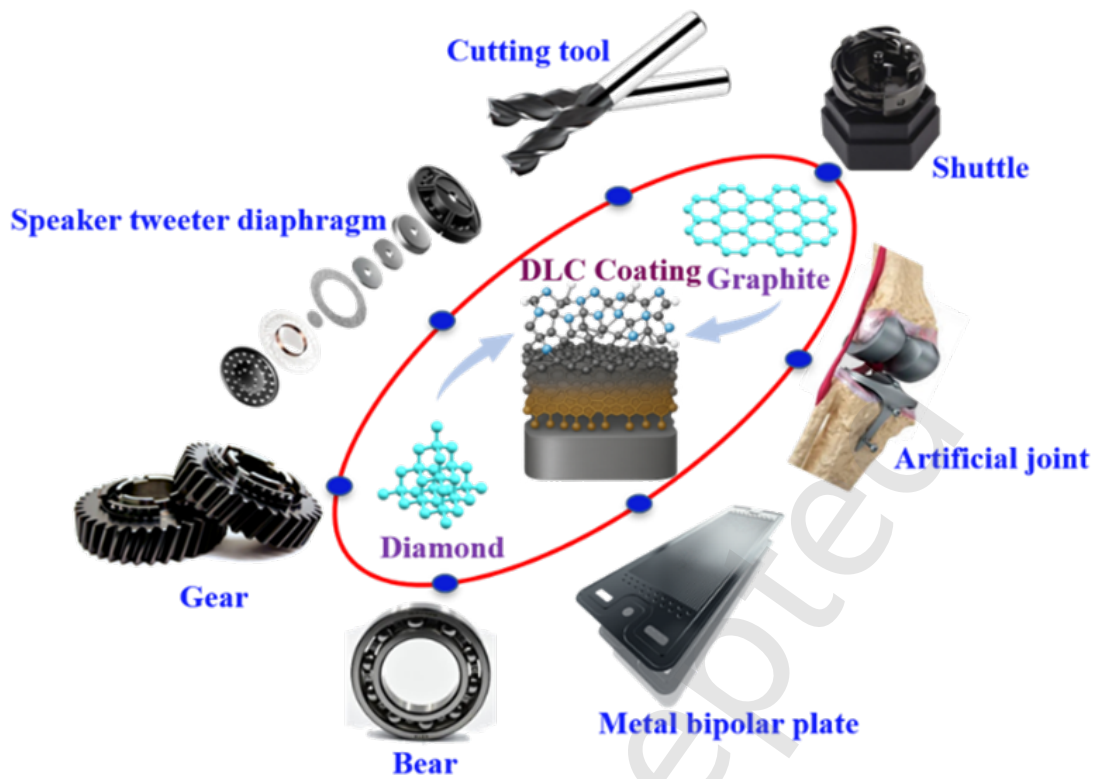


Fig. 1 The extensive applications of DLC coatings.

Despite the numerous advantages of DLC coatings in terms of mechanical strength and tribological performance, their application in extreme environments, such as those encountered in aerospace, still faces several critical bottlenecks. Among these factors, inherent deficiencies such as insufficient coating adhesion, low toughness, and high brittleness have significantly constrained the broader engineering application and widespread adoption of DLC coatings. The high residual stress within the coating [40-43], coupled with the chemical incompatibility between the coating and the substrate [44], represents the principal cause of interfacial adhesion degradation, which in turn predisposes the coating to cracking, delamination, and related failure modes. Moreover, although the amorphous structure of DLC coatings endows them with high hardness, it concurrently exacerbates their inherent brittleness, rendering them highly susceptible to brittle fracture under external impact or shear stresses, thereby undermining service stability and reliability [45-47]. The tribological behavior of DLC coatings is further complicated by their pronounced sensitivity to service environments, where factors such as

atmospheric composition, humidity levels, and temperature fluctuations can induce alterations in the chemical state of the coating surface, thereby influencing both the coefficient of friction and the underlying wear mechanisms [48-53]. For example, in high-humidity environments, moisture adsorption can facilitate the formation of a lubricating film on the coating surface, thereby effectively lowering the coefficient of friction [50, 51]. Conversely, in oxidizing atmospheres, the carbon structure at the surface is susceptible to oxidation, which leads to an increased coefficient of friction and accelerated wear [48, 49, 52, 53]. In addition, the increase or decrease of ambient temperature may also trigger the structural reconfiguration or degradation of coating performance [6, 7, 54, 55]. Therefore, in the material design and application of DLC coatings, it is essential to comprehensively consider their structural stability, interfacial adhesion, and environmental adaptability to ensure long-term stable performance and high reliability under complex and variable operating conditions.

To overcome the challenges of insufficient adhesion, inherent brittleness, and environmental sensitivity of DLC coatings under extreme operating conditions, numerous performance regulation strategies have been developed. Current research mainly focuses on three representative approaches: multilayer architecture design [56-60], heterogeneous element doping [61-65], surface topography engineering [66-70], and solid-liquid lubrication regulation. Among them, multilayer architectures can effectively relieve residual stress, enhance interfacial bonding strength, and improve crack resistance through the introduction of transition or gradient layers [71-74]. Heterogeneous element doping enables regulation of the sp^2/sp^3 bonding structure and microstructural characteristics of DLC coatings, thereby improving mechanical properties, tribological performance, thermal stability, and environmental adaptability [75-81]. In addition, surface topography engineering can regulate interfacial contact states and lubricating film formation, thus enhancing friction-reduction and wear-resistance capabilities [69, 82-87]. Solid-liquid lubrication regulation enhances tribological performance by promoting stable low-shear films and adaptive lubrication, thereby reducing friction and wear under extreme conditions. These strategies provide important pathways for improving the

reliability and service performance of DLC coatings in demanding aerospace and precision engineering applications. Detailed discussions of these regulation mechanisms are presented in the following sections.

This article addresses the critical challenge of insufficient tribological performance of DLC coatings on essential aerospace components operating under extreme conditions. As illustrated in Fig.2, initially, it analyzes the pronounced sensitivity of various DLC coating types to environmental factors such as temperature, atmosphere, and humidity, elucidating the underlying mechanisms of their impact. Then, it reviews the prevailing strategies for optimizing the tribological behavior of DLC coatings, encompassing structural design, compositional regulation, and surface modification, along with their fundamental mechanisms. Next, a comprehensive overview of the current status and engineering challenges related to the application of DLC coatings in typical aerospace components is then provided. Finally, the article concludes by anticipating future development trends in DLC coatings within aerospace technology and proposing several promising research directions, including the design of novel multilayer coating architectures, the application of nanoscale doping techniques, and the exploration of intelligent, responsive surface engineering strategies. These insights aim to offer valuable guidance for enhancing the tribological reliability of aerospace systems operating under complex service conditions.

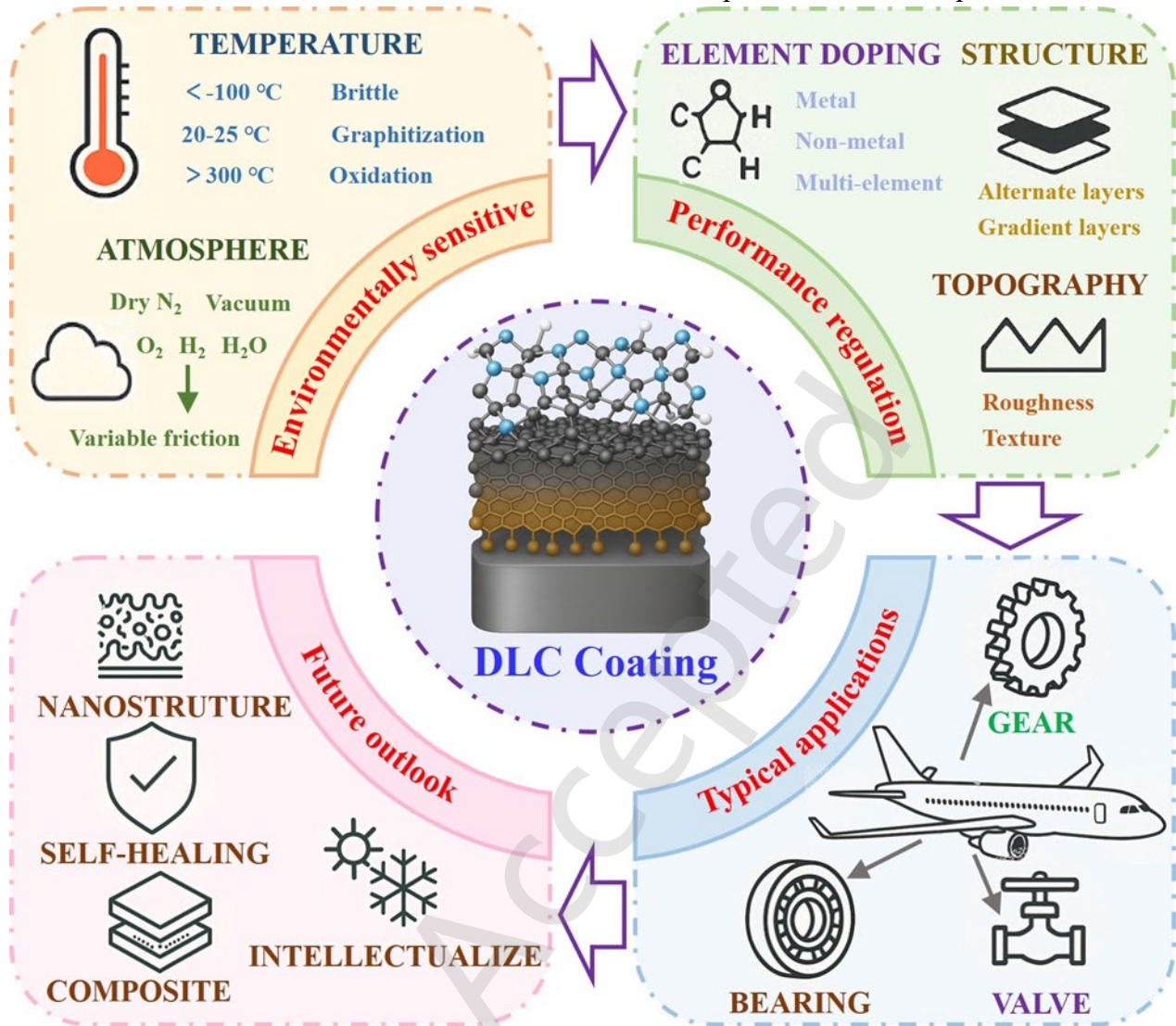


Fig. 2 Overview of the article structure

2 Strong environmental dependence of the tribological properties of DLC coatings

As an advanced solid lubricant material, DLC coatings exhibit significant variations and highly complex evolution patterns in their tribological performance and wear mechanisms under different service environments, including but not limited to ambient atmosphere, vacuum, high humidity, extreme temperatures, and corrosive media [48]. Particularly in aerospace extreme conditions, DLC coatings must withstand multiple challenges such as ultra-high/low temperature cycling, atomic oxygen erosion, high-vacuum cold welding, and fretting wear, demonstrating strongly environment-

dependent tribological behavior. This characteristic primarily stems from the dynamic evolution of coating surface chemistry, tribochemical reaction products, and interfacial contact characteristics during service. A fundamental understanding of environmental effects on DLC coating tribology and their structure-performance relationships is crucial not only for revealing the intrinsic wear mechanisms, but also for providing theoretical foundations and engineering guidance for composition design, structural optimization, and performance regulation under specific operating conditions. Consequently, prominent research groups worldwide have conducted systematic investigations on DLC coating tribological behavior in multi-field coupled environments, achieving groundbreaking progress in interfacial reaction mechanisms, have not only considerably enriched and expanded the theoretical framework of solid lubrication, but also laid the foundation for rigorous scientific research in the field of engineering applications of DLC coatings in the aerospace industry [88].

2.1 Temperature-dependent tribological properties of DLC coatings

Ambient temperature plays a critical role in governing the tribological behavior of DLC coatings. Under varying thermal conditions, different types of DLC coatings exhibit marked disparities in interfacial reactivity, structural stability, and friction and wear characteristics, highlighting their pronounced temperature-sensitive tribological nature [89, 90]. Despite substantial research efforts in this field, the failure mechanisms of DLC coatings under high-temperature environments remain a subject of ongoing debate. The extant literature has focused predominantly on the potential degradation modes, including high-temperature oxidation, structural destabilization, and graphitization. Nevertheless, the fundamental mechanisms underpinning these phenomena, along with their coupled and multifunctional effects, have yet to be fully elucidated.

Specifically, conventional undoped amorphous carbon (a-C) coatings exhibit marked degradation in performance under high-temperature operating conditions [91, 92]. Once the ambient temperature surpasses the critical threshold of 120 °C, the intrinsic wear-resistant mechanisms effective under ambient conditions begin to collapse. More critically, upon exposure to high-temperature annealing in

oxygen-rich atmospheres, these coatings not only undergo significant structural destabilization but also initiate severe oxidative degradation processes, culminating in a substantial decline in tribological performance. This intrinsic vulnerability fundamentally constrains the applicability of pure carbon-based DLC coatings in high-temperature oxidative environments [93, 94]. Moreover, given the pivotal role of surface-adsorbed water films in enabling ultralow friction coefficients and superior anti-wear performance, the operational window of such coatings is inherently restricted to ambient temperatures with moderate humidity. Interestingly, the wear behavior of DLC coatings displays an anomalous relationship with the hardness of the counterface materials. Despite the significantly lower hardness of 319 aluminum alloy compared to traditional hard materials such as tungsten carbide and sapphire, it induces more pronounced wear on the DLC surface, a phenomenon that becomes more severe at elevated temperatures. To elucidate this counterintuitive observation, researchers have proposed a composite mechanistic model incorporating both two-body and three-body abrasive wear, which provides a theoretical basis for understanding the failure mechanisms of DLC coatings under high-temperature service conditions [93-95].

In the investigation of the high-temperature tribological behavior of tetrahedral amorphous carbon (ta-C) coatings, researchers systematically examined the evolution of frictional performance for ta-C films deposited on 319 aluminum alloy substrates. The findings indicated a significant negative temperature dependence in the steady-state friction coefficient. As the ambient temperature increased from 25 °C to 100 °C, the coefficient exhibited a pronounced decline from 0.39 to 0.20. Further temperature elevation to 400 °C led to a further decrease to approximately 0.15. This behavior is in stark contrast to the typical degradation observed in conventional DLC coatings at elevated temperatures. Detailed analysis attributes the exceptional high-temperature stability of ta-C coatings chiefly to their elevated sp^3 bond proportion, which bestows robust structural integrity under extreme thermodynamic conditions. More notably, the friction-induced local phase transformation from sp^3 to sp^2 during sliding contact leads to the formation of a graphitized transfer layer at the interface. This

self-lubricating layer, generated in situ, plays a pivotal role in achieving and sustaining an ultra-low friction state [96, 97].

As demonstrated in Fig.3(a), existing research on the high-temperature tribological behavior of hydrogenated diamond-like carbon (H-DLC) coatings has revealed that these coatings exhibit exceptional thermal stability, maintaining both structural integrity and tribological performance at temperatures reaching up to 500 °C. Moreover, they retain an ultralow coefficient of friction (below 0.05) and demonstrate excellent wear resistance under such extreme conditions. In comparison to hydrogen-free DLC coatings, H-DLC shows markedly superior lubricating properties at elevated temperatures. This advantage arises from the stabilizing effect of hydrogen on the carbon network during frictional contact, as well as the formation of a dense, continuous lubricant film on the coating surface. Under high-temperature sliding conditions, H-DLC coatings facilitate the development of a uniform and stable carbon-rich transfer film or friction-induced layer on the counterface, which effectively suppresses oxidative wear mechanisms, significantly retards the degradation process, and ensures prolonged protection of the substrate. Importantly, as the frictional temperature increases, the tribological behavior of H-DLC coatings undergoes a progressive transformation from an adhesion-dominated mechanism to a tribofilm-dominated one. This transition enables a shift from severe wear to mild wear regimes, thereby further enhancing the coating's friction-reducing and anti-wear capabilities under extreme service environments [38, 98].

In recent years, addressing the pronounced temperature dependence of the tribological properties of DLC coatings through compositional engineering has emerged as a prominent research frontier. A representative approach involves doping with metallic elements such as Cr and Ti, both of which exhibit strong chemical affinity for carbon. Although such doping may slightly reduce the intrinsic hardness of the coating, the induced structural reconfiguration, coupled with the formation of a protective carbon-metal oxide barrier layer at the frictional interface, significantly enhances the coating's wear resistance and frictional stability under medium-to-high temperature conditions (e.g.,

250 °C and 300 °C) [99-101]. Similarly, as illustrated in Fig.3(b), Si doping demonstrates a notable capacity to regulate high-temperature tribological behavior. Within the amorphous carbon matrix, Si is present as disordered tetrahedral silicon carbide phases, which not only enhance the toughness of the coating but also promote a structural transformation toward a diamond-like configuration. This transformation contributes to increased hardness and enables stable low-friction performance at temperatures as high as 500 °C. This superior performance is primarily attributed to the improved structural stability resulting from the increased sp^3 bonding fraction, as well as to the thermally induced formation of a SiO_2 -rich protective film at the counterface, which acts as an effective barrier to wear and oxidation, thereby enabling sustained lubrication under extreme thermal conditions [102]. Furthermore, as shown in Fig.3(c-d), both MoS_2 and B_4C compounds have independently demonstrated remarkable efficacy in enhancing the high-temperature tribological performance of DLC coatings, albeit through distinct mechanisms. MoS_2 doping notably improves the thermal stability of the coatings, enabling the attainment of ultralow friction at 250 °C, attributed to the surface-layer precipitation of the MoS_2 phase. As the temperature increases to 350 °C, a $MoS_{2-x}O_x$ solid solution forms at the sliding interface, maintaining the low-friction regime. Even at elevated temperatures of 450 °C, coatings with high MoS_2 doping levels exhibit stable operation, benefitting from the sustained lubricating action of the $MoS_{2-x}O_x$ solid solution [103]. Concurrently, Moderate B_4C doping elevates the optimal lubrication temperature from 300 °C, typical of undoped DLC, to 500 °C, at which an ultralow friction coefficient as low as 0.04 is achieved. This exceptional performance is primarily attributed to the shear lubrication effect induced by temperature-driven surface graphitization, in conjunction with the formation of a solid lubricating film composed of boron oxide-enriched graphitized debris within the wear track [104].

In the currently limited body of research concerning the sensitivity of tribological performance variations of DLC coatings under cryogenic conditions, scholars have primarily sought to elucidate the key factors influencing the evolution of frictional behavior in ambient air atmospheres by analyzing

the tribological responses and wear scar morphologies of multilayer DLC coatings. It has been found that the formation of friction-induced transfer layers, the progressive graphitization of the coating surface, and the passivation of surface dangling bonds by hydrogen atoms constitute the fundamental mechanisms governing the observed changes in tribological behavior. Under room temperature conditions, the degree of surface graphitization is markedly higher compared to cryogenic environments. This can be attributed to the accelerated formation of transfer films at elevated temperatures, which facilitates the rapid establishment of a stable lubricating interface during the running-in stage, thereby enabling the attainment of ultralow friction coefficients. Nevertheless, continued graphitization may lead to a decline in both hardness and elastic modulus of the coating, resulting in more pronounced surface damage. In contrast, cryogenic environments significantly suppress both the graphitization rate and the generation of transfer films. Although a slight increase in friction coefficient is observed at lower temperatures, the inherently high sp^2 -hybridized carbon content within the DLC matrix ensures the preservation of favorable lubricating characteristics. Moreover, the saturation of surface dangling bonds in hydrogenated DLC (a-C:H) coatings by hydrogen atoms is also recognized as a critical factor in sustaining low-friction behavior. The low-temperature environment enhances the bond energy of C-H and C-C chemical bonds, thereby inhibiting interfacial bonding reactions between sliding counterparts. This suppresses surface chemical reactions and adhesion phenomena, fundamentally improving the tribological stability of the coating under cryogenic conditions [105]. Notably, with the advancement of research, investigations into the low-temperature tribological evolution of hydrogenated DLC coatings have progressively extended to multi-element doped systems. Against this backdrop, as illustrated in Fig.3(e), the introduction of dopant elements such as Al, Cr, and Si into a-C:H coatings has been shown to result in a more intricate, stage-wise evolution of tribological behavior under cryogenic conditions. As the temperature decreases, both shear-induced graphitization and the mobility of hydrogen atoms are markedly suppressed. Consequently, the frictional behavior transitions from an ultralow-friction regime dominated by shear-

induced graphitization to successive stages dominated by abrasive wear, adhesion of wear debris, and brittle fracture of the coating. When the environmental temperature falls to the extremely low range below -130°C , the initiation and propagation of cracks become more pronounced, accompanied by interlayer delamination, which leads to a significant deterioration in tribological performance [54].

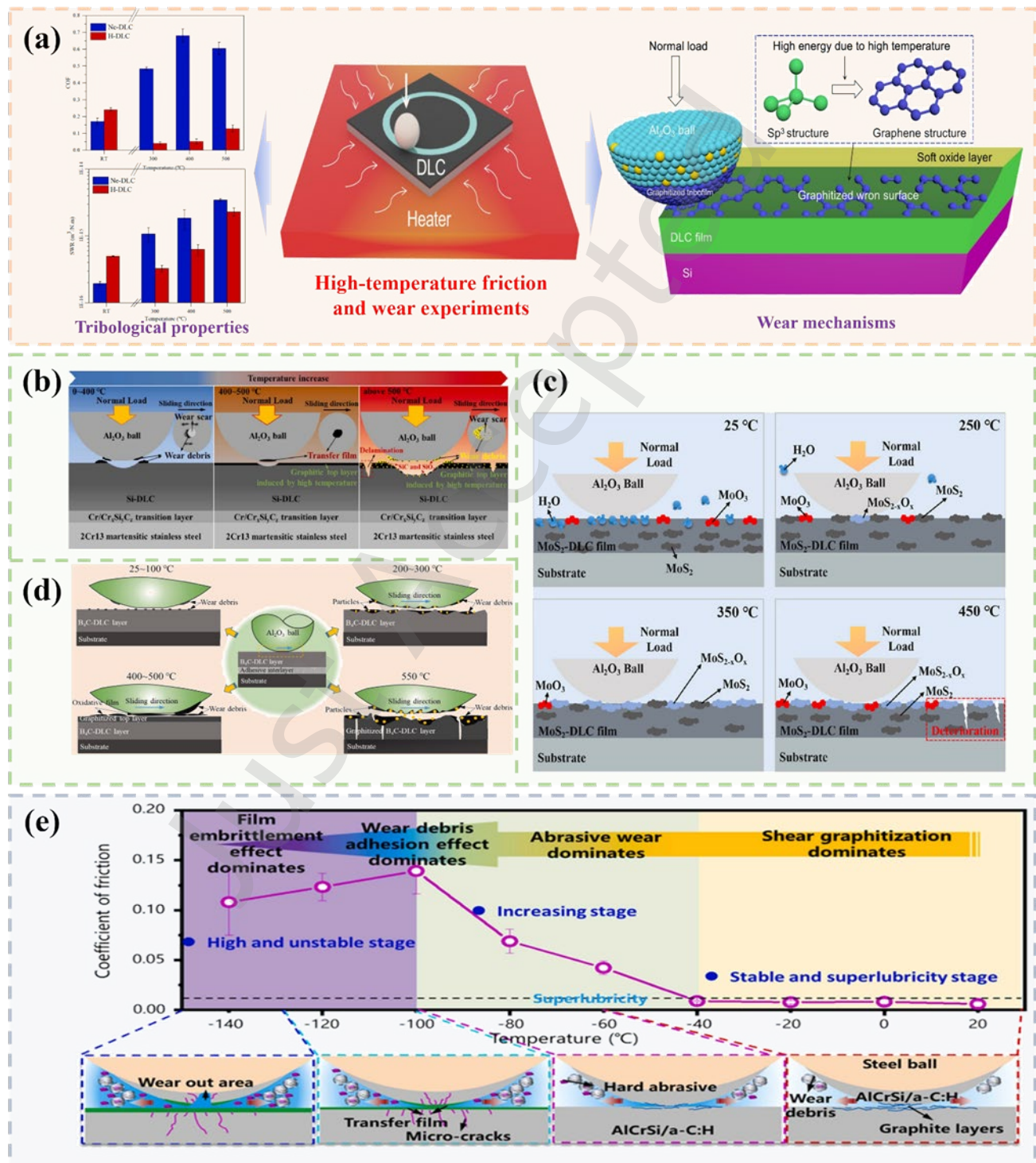


Fig. 3 (a) High-temperature tribological properties and wear mechanisms of hydrogenated DLC coatings [38, 98]. (b) Schematic diagram of high-temperature wear mechanisms of Si-DLC coatings [102]. (c) Schematic diagram of high-temperature wear mechanisms of MoS₂-DLC coatings [103]. (d) Schematic diagram of high-temperature wear mechanisms of B₄C-DLC coatings [104]. (e) Illustration of the carbon film friction evolution at low-temperatures [54].

Importantly, recent studies have further elucidated the intrinsic role of hydrogen in modulating the tribological behavior of DLC coatings under cryogenic conditions. It has been demonstrated that hydrogenated amorphous carbon films exhibit a characteristic hydrogen-mediated sub-Arrhenius frictional behavior in the deep cryogenic regime, while achieving stable ultralow friction coefficients across a broad temperature range of 103 to 348 K. This performance is primarily attributed to the key role of hydrogen in inducing a mild structural ordering transformation within the DLC matrix during friction. The temperature-dependent nature of this structural evolution can be accurately described by an Arrhenius model modified by quantum tunneling effects, which reflects the quantum delocalization characteristics of hydrogen in tribochemical reactions [106]. These findings not only provide a theoretical foundation for understanding the tribological response of DLC coatings under extreme temperatures but also offer valuable guidance for the rational design of reliable lubricating interface materials suitable for cryogenic environments.

Overall, the temperature-dependent of the tribological properties of DLC coatings reveals a significant mutual coupling feature among structure, composition, and interfacial characteristics, exhibiting a discernible stage-wise evolution mechanism across diverse thermal regimes. At elevated temperatures, doping elements significantly enhance the thermal resistance and high-temperature lubrication stability of DLC coatings by inducing structural reconstruction, promoting the precipitation of lubricious phases, or facilitating the formation of stable protective films at the frictional interface, such as metal oxides, SiO₂, and MoS_{2-x}O_x. In contrast, under low and cryogenic temperature conditions, the frictional behavior of the coatings is primarily constrained by the graphitization effect, the

migration kinetics of hydrogen atoms, and the suppression of interfacial chemical reactivity, among which the quantum-dominated role of hydrogen atoms emerges as the key factor ensuring the coatings' frictional stability at low temperatures. It is imperative to underscore that the capacity of DLC coatings to sustain superior tribological performance across a broad temperature spectrum hinges upon a multi-scale, multi-mechanism synergistic regulation strategy. This encompasses the precise selection and stoichiometric optimization of dopant elements, the rational architectural design of microstructural ordering, and the dynamic modulation of friction-induced interfacial films. Particularly under extreme thermal conditions, the stability of their frictional behavior not only epitomizes the intrinsic structural robustness of the material but also underscores the intricate challenges associated with governing interfacial tribo-responses. Consequently, a fundamental elucidation of the structural evolution mechanisms and interfacial reaction pathways of DLC coatings under varying thermal regimes constitutes a critical prerequisite for ensuring their operational reliability in demanding applications, including aerospace propulsion, deep-space exploration and cryogenic fluid management. Meanwhile, the tribological behavior of DLC coatings is governed by the dynamic coupling among composition, structure, tribochemical evolution, and environmental conditions, highlighting the necessity of establishing quantitative composition-structure-environment-performance relationships. Future investigations should prioritize unraveling the synergistic mechanisms of multi-elemental doping, probing the electronic structure modulation effects at tribo-interfaces, and advancing quantum-scale modeling of friction dynamics. In addition, the integration of multiscale simulations, machine learning-assisted materials design, and advanced in situ characterization techniques is expected to facilitate the development of predictive frameworks for the rational design and performance optimization of DLC coatings under extreme environments. Such endeavors are pivotal in driving paradigm-shifting theoretical advancements and facilitating the practical deployment of temperature-adaptive lubricious materials, thereby laying a robust foundation for the next generation of advanced tribo-functional coatings.

2.2 Strong sensitivity of tribological performance of DLC coatings to service gas atmospheres

In addition to temperature, the gaseous atmosphere present in the service environment plays a critical and multifactorial role in governing the tribological performance of DLC coatings. In aerospace applications, different gaseous environments are closely associated with specific operational scenarios. For example, oxygen-rich atmospheres are commonly encountered in liquid oxygen valve systems and propulsion components, while high-vacuum environments are representative of low-Earth orbit and deep-space spacecraft conditions. In contrast, air atmospheres are mainly related to atmospheric flight environments, whereas nitrogen atmospheres are often used as inert protective conditions during ground testing or sealed operating systems. Variations in gas composition under these environments can profoundly influence the tribological behavior of DLC coatings by modulating interfacial chemical reactions, friction-induced structural transformations, and tribochemical processes [107]. Under extreme aerospace environments, such as oxygen-rich atmospheres and high vacuum conditions, the evolution of surface-active species, the formation of tribochemical products, and their spatial distribution are significantly altered, thereby directly affecting lubrication behavior and wear resistance. Moreover, the type and partial pressure of surrounding gases can regulate the formation and stability of boundary lubrication films and may either promote or suppress carbon network rearrangements, ultimately influencing the structural stability and long-term reliability of DLC coatings. Therefore, the gaseous atmosphere should be regarded as a key environmental factor in understanding the tribological mechanisms and engineering applications of DLC coatings in aerospace systems.

Several scholars have employed a combination of reactive force field molecular dynamics simulations and density functional theory calculations to investigate the tribological behavior and interfacial reaction mechanisms of hydrogen-free DLC coatings under various gaseous environments, as illustrated in Fig.4(a). In a nitrogen atmosphere, the introduction of nitrogen molecules effectively suppresses the formation of C-C covalent bonds at the contact interfaces of the frictional counterparts,

thereby delaying the rapid rise of the initial friction force [108]. However, as sliding progresses, the infiltration and diffusion of nitrogen molecules into the interface induce pronounced interfacial mechanical mixing effects. The resulting high shear stresses detrimentally influence the tribological performance of the coating [109]. In contrast, under oxygen-rich conditions, oxygen atoms exhibit significant interfacial enrichment while their penetration into the coating remains minimal. These enriched interfacial oxygen atoms contribute to the realization of an ultralow-friction regime by facilitating the formation of a gas-dynamic lubrication-like layer and reducing the interfacial carbon atom density. Moreover, the study emphasizes that within this system, the conventionally proposed mechanisms of graphitization and surface passivation do not dominate the frictional behavior [52, 110]. Compared with hydrogenated DLC coatings, hydrogen-free DLC exhibits fundamentally different tribological mechanisms under different gaseous environments. For hydrogen-free DLC, friction behavior is mainly governed by interfacial dangling-bond activity, tribochemical passivation, and friction-induced graphitization. In contrast, hydrogenated DLC coatings rely more strongly on hydrogen termination effects, transfer-film evolution, and suppression of interfacial adhesion. Under oxygen-rich atmospheres, hydrogen-free DLC can achieve partial passivation through the formation of oxygen-containing functional groups, thereby reducing interfacial adhesion and promoting low-friction behavior. However, for hydrogenated DLC coatings, oxygen tends to consume surface hydrogen atoms and destroy the originally passivated interface, resulting in re-exposure of active carbon dangling bonds and deterioration of tribological performance. In a hydrogen atmosphere, the a-C coating undergoes intense plastic deformation during sliding, accompanied by substantial energy release and a sharp rise in interfacial temperature, which progressively promotes the graphitization of the coating. However, the friction force continues to increase throughout the process. Further investigations reveal that effective hydrogen passivation is difficult to achieve for a-C coatings in an H₂ environment. This observation indicates that neither graphitization nor hydrogen passivation is sufficient to effectively reduce friction in such conditions. On the contrary, elevated temperatures

facilitate the diffusion of H₂ molecules into the coating, thereby intensifying mechanical mixing at the interface, increasing the contact density of carbon atoms, and ultimately resulting in a higher steady-state friction force. Moreover, the study highlights that within this system, the conventionally proposed mechanisms of graphitization and surface passivation do not serve as the dominant factors governing the tribological behavior [109]. Regarding the CO₂ atmosphere, the physical adsorption of CO₂ molecules at the friction interface markedly decreases the initial friction force. At the steady-state stage, the persistently low friction coefficient is attributed to the formation of numerous C-O terminal functional groups, generated through a synergistic interplay between physical and chemical adsorption. These functional groups effectively mitigate friction-induced covalent bond ruptures [110]. Further interfacial bonding simulations demonstrate that the DLC surface can approach complete passivation when the coverage of oxygen or nitrogen exceeds 0.75. Nevertheless, compared with nitrogen, the C-O bonds formed in an oxygen atmosphere exhibit lower bond strength and stronger interatomic repulsive interactions at the interface. This leads to reduced friction coefficients, smoother contact interfaces, and a diminished number of interfacial bonds, thereby providing a theoretical foundation for the superior tribological performance observed under oxygen compared to nitrogen environments [111, 112].

In addition, many fruitful advances have been achieved in elucidating the influence of gaseous environments on the tribological behavior of DLC coatings from an experimental perspective. Taking a-C:H coatings as a representative example, their tribological performance deteriorates markedly in oxygen-rich atmospheres. As illustrated in Fig.4(b), this degradation is primarily attributed to a series of tribochemical reactions occurring at the sliding interface. Upon exposure to oxygen-containing environments, surface hydrogen atoms preferentially react with oxygen species, resulting in the formation of oxygen-containing functional groups such as water molecules and hydroxyls [36]. This process induces a dual deleterious effect: on the one hand, the loss of hydrogen atoms leads to the re-exposure of formerly passivated carbon dangling bonds, thereby enhancing the surface chemical

activity; on the other hand, the formation of polar oxygenated groups alters the surface chemical state and interfacial interaction characteristics. At the microscopic level, these newly exposed carbon dangling bonds engage in strong chemical interactions with the counterface material, significantly increasing interfacial adhesion [105, 113, 114]. Although both hydrogen-free DLC and hydrogenated DLC are strongly influenced by atmospheric environments, their dominant friction mechanisms exhibit clear differences. Under inert atmospheres such as nitrogen, hydrogenated DLC coatings can achieve ultralow friction owing to hydrogen-passivated surfaces and interfacial dipole repulsion effects, whereas hydrogen-free DLC mainly depends on environmental molecular adsorption and suppression of direct C-C bond formation. Under vacuum conditions, both systems suffer from the absence of environmental passivation. However, hydrogen-free DLC coatings are more susceptible to severe adhesive interactions and brittle spalling due to the large number of unsaturated dangling bonds. By contrast, hydrogenated DLC coatings can still maintain relatively low friction because residual hydrogen atoms facilitate the formation of a polymer-like hydrocarbon transfer film with low shear strength [115]. In recent years, increasing attention has been devoted to the tribological adaptability of DLC coatings in oxidative environments. The incorporation of tungsten (W) has emerged as an effective strategy, profoundly influencing both the bonding configuration and graphitization degree of the coatings. More importantly, it mitigates the adverse interfacial chemical reactions arising from hydrogen loss or carbon bond exposure. Specifically, the introduction of W atoms can lead to the localized formation of WC or W-C composite phases, which contribute to structural densification and enhanced oxidation resistance. Under frictional conditions, these phases may undergo tribo-induced oxidation to produce tungsten oxides such as WO_3 . These oxides exhibit low shear strength and excellent thermal stability, facilitating the formation of a nanometric boundary film with intrinsic self-lubricating characteristics at the sliding interface. This boundary film effectively reduces interfacial adhesion and shear stress, while simultaneously serving as a barrier to oxygen diffusion, thereby

retarding direct oxidation of the coating. Consequently, long-term tribological stability can be achieved even under oxygen-rich conditions [116-118].

In contrast, under inert gas atmospheres such as nitrogen, hydrogenated DLC coatings can achieve extremely low coefficients of friction, reaching values of approximately 0.005 under a normal load of 2 N [119, 120], as shown in Fig.4(c). This remarkable tribological performance is primarily attributed to the high concentration of hydrogen both within the bulk and on the surface of the coating. The abundant hydrogen atoms effectively passivate reactive σ -type carbon dangling bonds, thereby suppressing the likelihood of strong interfacial adsorption with the counterface material during sliding contact [119, 121, 122]. Furthermore, the C-H bonds formed at the sliding interface are highly chemically inert, which significantly reduces the adhesive interaction between the carbon atoms of the coating and the mating surface. This inert interfacial chemistry leads to a substantial decrease in interfacial shear strength and contributes to the pronounced reduction in friction. More importantly, the electronic structure of the C-H bond further reinforces its anti-adhesive properties. When the free electrons of hydrogen atoms pair with the unbonded electrons associated with carbon dangling bonds, a redistribution of charge density occurs. This redistribution shifts the electron density toward the hydrogen atoms, positioning the positively charged hydrogen nuclei closer to the surface of the coating. As a result, directional C-H and H-C dipole repulsions are generated at the sliding interface. These electrostatic repulsions effectively weaken van der Waals forces and transient electrostatic attractions between the contact surfaces, thereby reducing interfacial adsorption and energy barriers. Collectively, these effects endow hydrogenated DLC coatings with superior friction-reducing capabilities under inert environmental conditions [119, 123].

Under vacuum conditions, the absence of environmental mediators causes hydrogen-free DLC coatings to continuously generate a substantial number of unsaturated carbon dangling bonds during frictional contact. These highly reactive sites markedly elevate the surface free energy of the film and facilitate strong chemisorptive interactions with the counterface material. Concurrently, adhesive

forces within the tribological interface become significantly pronounced at the atomic scale, resulting in a substantial enhancement of interfacial bonding strength compared to ambient atmospheric conditions [48, 124]. It is noteworthy that, as illustrated in Fig.4(d), the passivation of the active sites by environmental molecules under vacuum conditions is completely absent, which shifts the wear mechanism from plastic deformation, which is dominant in conventional atmospheric environments, to a brittle spalling-dominated failure mode. This shift directly leads to the hydrogen-free DLC coatings presenting high friction coefficients and wear rates under vacuum conditions. For a-C:H coatings, the surface dangling bonds are effectively passivated due to the saturation by hydrogen atoms [124-126], accompanied by the formation of an ultrathin, polymer-like hydrocarbon transfer film on the surface. The transfer film has a typical lamellar structure, which is highly susceptible to directional alignment and low-resistance shear along the sliding direction under the action of shear force, thus conferring excellent friction reduction properties to the hydrogenated DLC coating. This structural property allows hydrogenated DLC coatings to maintain a relatively low coefficient of friction under vacuum, but their wear resistance is often limited by the stability of the transfer film [127]. The hydrogen content, in turn, is a key parameter that determines the tribological performance of DLC coatings under vacuum, and the level of hydrogen content directly affects the degree of saturation of the suspended bonds on the surface of the coatings and the ability of the formation of the transfer film, which then dominates its interfacial lubrication mechanism. When the hydrogen content is relatively low, the number of saturated bonds in the coating decreases, the number of surface-active sites increases, and the direct contact and atomic-level adhesion of C-C strong bonds are very easy to occur on the surface of the coating under high vacuum due to the lack of sustained passivation of ambient molecules, thus significantly exacerbating the interfacial friction and wear process. More critically, such low-hydrogen DLC is difficult to form a stable and sustainable low-shear transition film during the friction process, and its intrinsic lubrication performance declines rapidly, which is manifested as a rapid failure of lubrication efficiency and a significant increase in the coefficient of friction, and even

results in the transformation of the wear mechanism to brittle spalling. Therefore, rational regulation of the hydrogen content of DLC coatings is one of the key strategies to enhance their service stability and tribological performance under high vacuum conditions [124, 128].

Besides, as illustrated in Fig. 4(e), the tribological behavior of DLC coatings is strongly influenced by atmospheric water vapor, whose tribochemical effects during sliding are closely related to those observed in oxygen-containing atmospheres. Under a normal load of 2 N, a-C:H coatings exhibit an extremely low coefficient of friction (COF) of approximately 0.06 at low relative humidity (14% RH), whereas a-C coatings show a much higher COF of about 0.19 under similar atmospheric conditions (17% RH). With increasing relative humidity, the COFs of both coatings gradually converge to approximately 0.15, and a similar trend is observed under a higher normal load of 5 N, indicating that the influence of atmospheric water vapor remains stable across different loading conditions. The superior low-friction behavior of a-C:H coatings under low-humidity atmospheres is mainly attributed to the formation of a graphitized tribofilm with low shear strength during sliding [129]. In contrast, the relatively high friction of a-C coatings originates primarily from strong adhesive interactions associated with unsaturated surface dangling bonds. As the atmospheric water vapor content increases, water molecules adsorb and penetrate into the sliding interface, suppressing direct solid-to-solid contact and altering the dominant friction mechanism. Under such conditions, capillary interactions gradually become dominant, while the absence of stable friction-induced carbonaceous tribolayers leads to similar frictional responses for both a-C:H and a-C coatings. Furthermore, oxygen molecules and water vapor in the atmosphere can jointly participate in interfacial tribochemical oxidation reactions during sliding, thereby accelerating tribological degradation and influencing the wear behavior of DLC coatings [129-133].

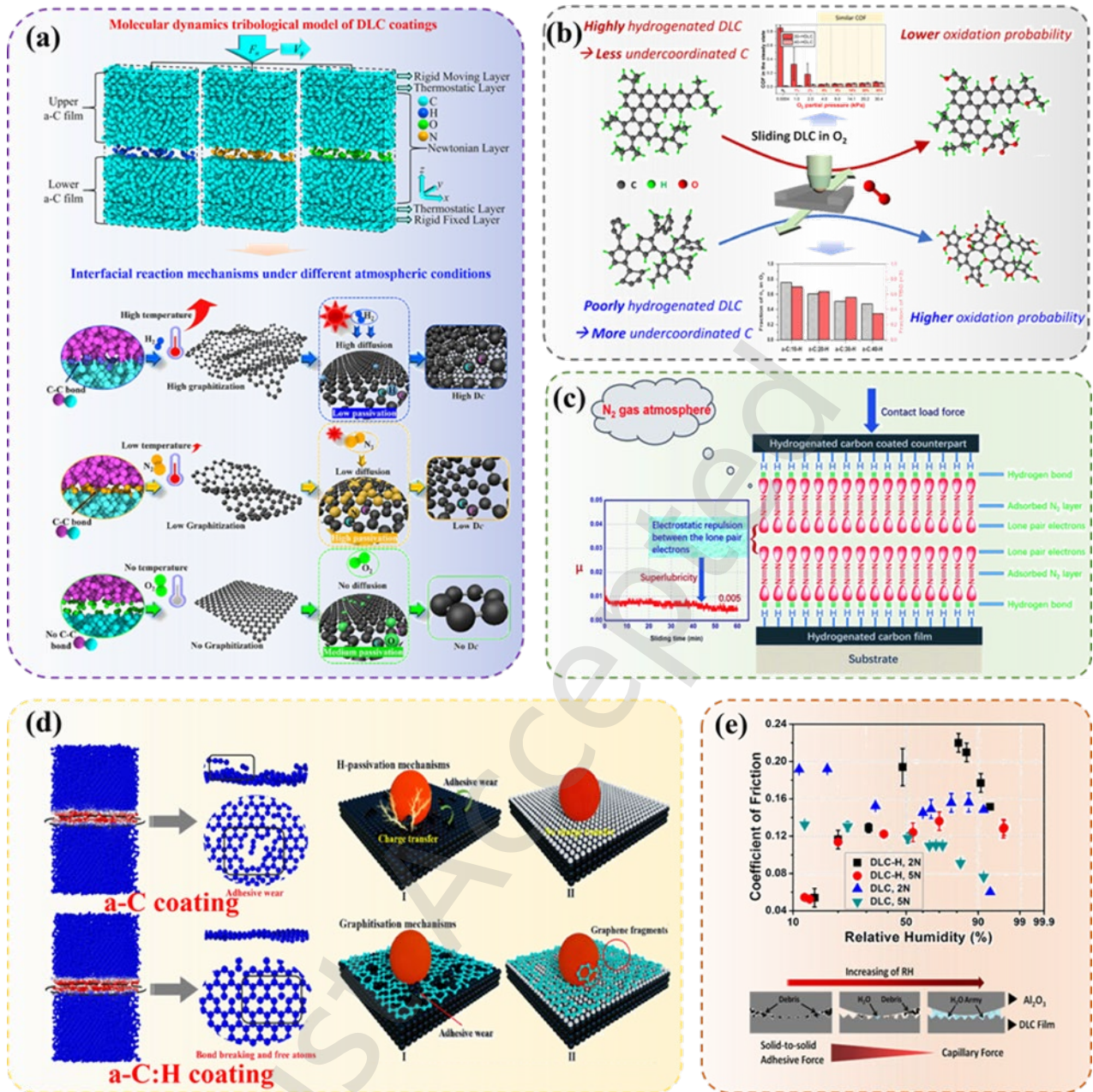


Fig. 4 (a) Frictional behavior and interfacial reaction mechanisms of DLC coatings under different atmospheres [109]. (b) Wear mechanism of a-C:H coatings in oxygen-rich environments [36]. (c) Schematic diagram of tribological mechanisms of a-C:H coatings in dry N₂ gas atmosphere [119]. (d) Wear mechanisms of a-C and a-C:H coatings under vacuum conditions [124]. (e) Tribological performance and wear mechanisms of DLC coatings under different humidity conditions [129].

In summary, the tribological behavior of DLC coatings exhibits a pronounced sensitivity to the surrounding gaseous environment, governed primarily by surface chemical state modulation and the

evolution of interfacial transfer films. Reactive gases, such as oxygen, have been observed to interact with unsaturated carbon bonds on the DLC surface, forming passivation layers. This process has been shown to effectively lower surface energy and chemical reactivity, thereby suppressing adhesion and wear. Conversely, under conditions of elevated vacuum, the augmented exposure of reactive carbon bonds impedes the formation of stable passivating structures, resulting in heightened interfacial energy and a substantial increase in the coefficient of friction. Despite their lack of participation in chemical surface reactions, inert gases such as nitrogen can exert a subtle influence on frictional behavior through their physical adsorption, which can alter the interfacial electronic state density. Furthermore, the gaseous atmosphere exerts a pivotal influence on the formation mechanism of transfer films during sliding. The presence of trace water vapor in ambient air promotes surface graphitization, leading to the generation of low-shear-strength lubricating films. Conversely, in vacuum environments, transfer film formation is driven by hydrogen-assisted surface reconstruction and the transformation of metastable carbon phases within the coating. Consequently, the gaseous environment exerts a critical influence on both the failure mechanisms and lubrication modes of DLC coatings by steering surface reaction pathways and interfacial structural evolution. This underscores the heightened demand for performance stability under complex operational conditions and offers a theoretical foundation for the structural optimization and environmental adaptability of DLC coatings.

3 Regulation strategies and mechanisms of tribological performance in DLC coatings

In order to surmount the critical bottleneck posed by the inadequate tribological performance of DLC coatings under extreme operating conditions in the aerospace sector, it is imperative to undertake comprehensive and systematic investigations into performance regulation strategies that address the demanding requirements of harsh service environments. As demonstrated in Fig.5, contemporary endeavors to enhance the performance of DLC coatings are predominantly concentrated in four primary domains: multilayer structures construction, heterogeneous elemental doping, surface morphology optimization and solid-liquid lubrication regulation. This section will focus on the

aforementioned four types of strategies, respectively, from the magnitude of tribological performance regulation and its intrinsic mechanism of two levels, to explore its mechanism and development potential, in order to provide theoretical support and design reference for the application of DLC coatings in the aerospace field of high reliability under complex service conditions.

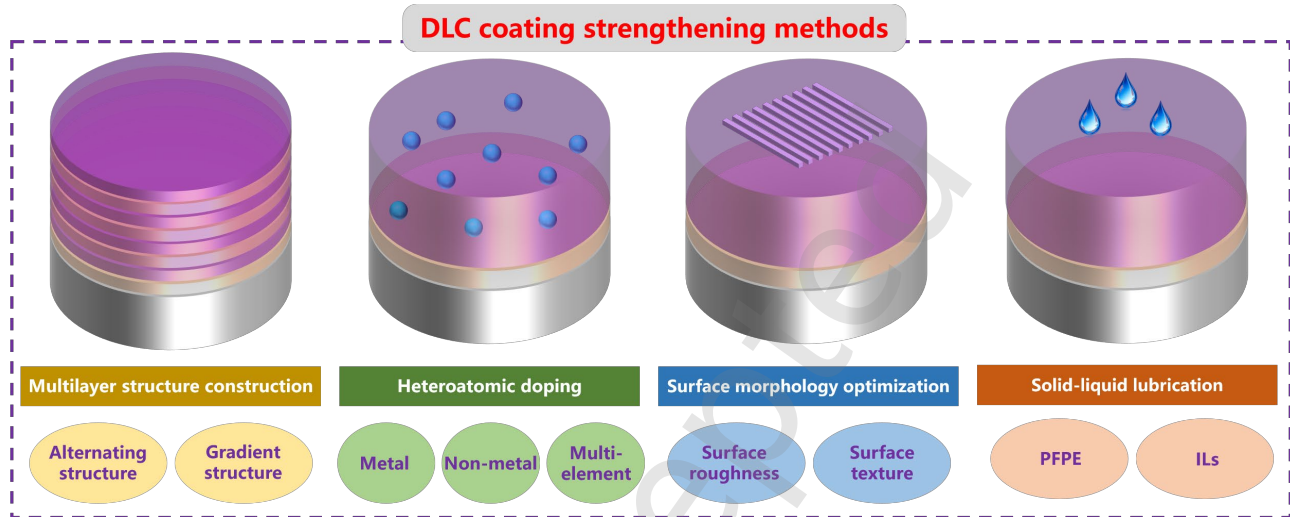


Fig. 5 DLC coating strengthening methods

3.1 Multilayer structure construction

The multilayer structural design of DLC coatings represents a performance enhancement strategy rooted in the hierarchical and gradient engineering of materials [57-59, 134-136]. The fundamental premise of this approach is to amplify advantageous properties and compensate for intrinsic limitations by regulating the arrangement of individual layers and leveraging interfacial coupling effects between functional sublayers [56, 79, 100]. Such multilayer architectures have been shown to mitigate internal stresses within the coating through stress redistribution mechanisms. Additionally, they have been demonstrated to enhance interfacial adhesion between the coating and the substrate via optimized interlayer design. Consequently, these architectures suppress failure modes such as microcrack propagation and delamination induced by high residual stress [137-139]. The “structure-for-performance” design paradigm offers a promising pathway to overcome the performance limitations of conventional monolithic DLC coatings. In the subsequent section, the architectural strategies and

reinforcement mechanisms of multilayer DLC coatings will be thoroughly examined, with a particular focus on alternating and gradient configurations.

3.1.1 Alternating structure

The conventional alternating structure design of DLC coatings is a performance modulation strategy based on the rational arrangement of hard and soft layers to optimize their performance and enhance their adaptability across a diverse range of operational conditions [140, 141]. In this configuration, hard layers impart high hardness and excellent wear resistance, ensuring stable performance under high loads and high-speed operations. Meanwhile, soft layers contribute enhanced toughness and cushioning capacity, which effectively reduce brittleness and help prevent crack initiation or structural failure caused by external impacts or elevated pressure. However, as illustrated in Fig.6, the underlying principle of this design is not limited to the classic alternation between hard and soft layers. It also encompasses a broader variety of heterogeneous interfacial combinations, such as hard-to-hard, soft-to-soft, and even metal-to-nonmetal interfaces [84, 137]. These extended configurations enable coordinated regulation of structure and properties across multiple scales, thereby offering a more versatile and robust approach to coating performance enhancement.

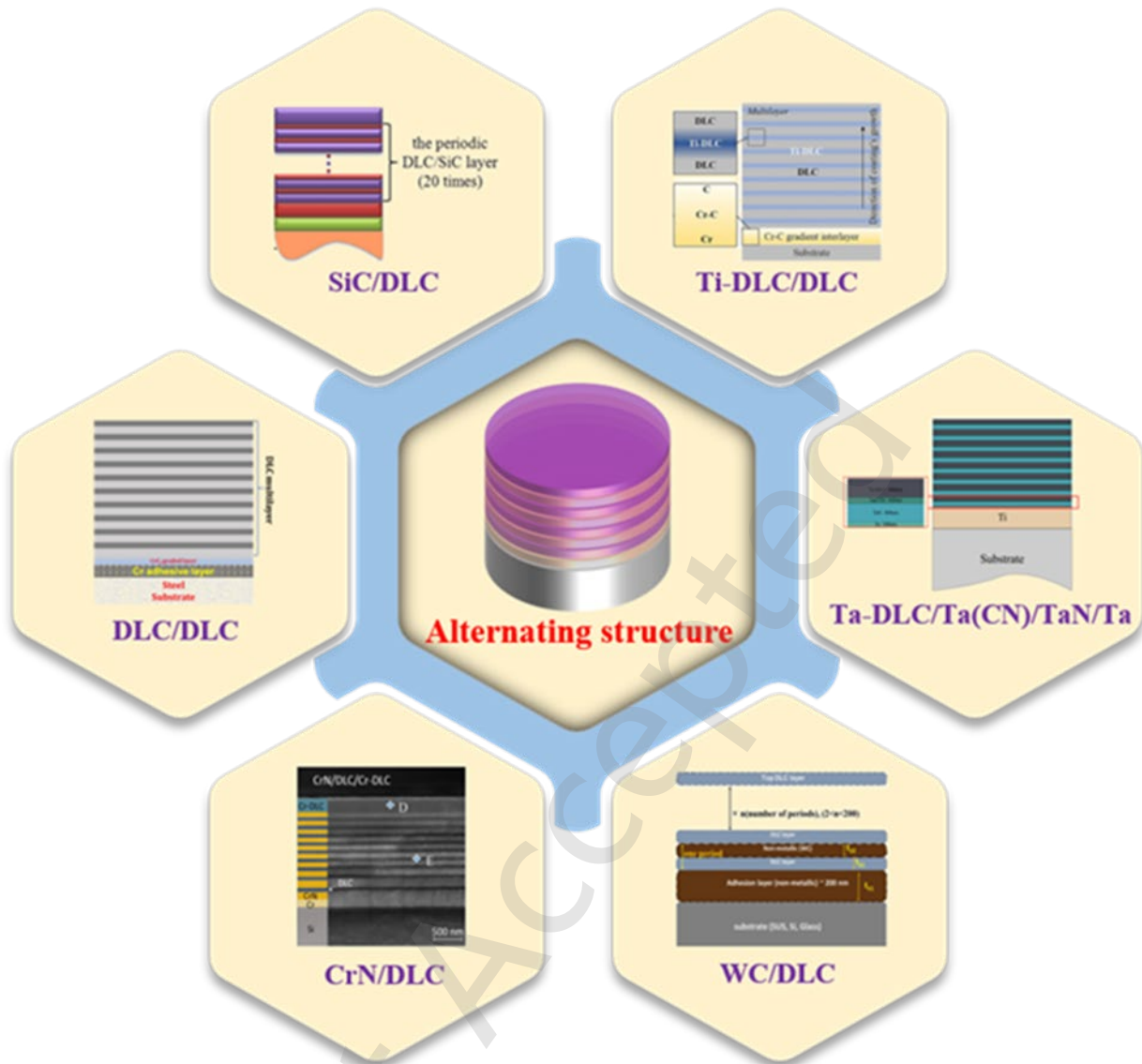


Fig. 6 Different types of alternating structures for DLC coatings [57, 73, 136, 142-144].

In several studies, Zhou et al. [142, 145-149] investigated the significant enhancement of the tribological properties of DLC coatings through the alternating soft and hard design of the carbon material itself. The findings in Fig.7(a) demonstrated that multilayer coatings with a soft top layer exhibited substantial improvements in wear resistance compared to single-layer DLC coatings. Specifically, the soft top layer facilitates the reduction of wear during the break-in period by forming a transport layer, which effectively diminishes overall wear [145]. Moreover, when investigating the impact of alternating layer parameters on tribological performance, it was found that as the thickness of the bilayer decreased with a soft-hard alternating ratio of 1:1, the coating's hardness, modulus, and

residual stress all increased. Further analysis revealed that under high contact stress, the coating achieved the minimum wear rate ($7.9 \times 10^{-8} \text{ mm}^3/\text{N} \cdot \text{m}$) when the number of alternating layers was 10 (see Fig.7(b)). These results highlight the importance of optimizing both the thickness and the number of alternating layers for enhancing the coating's wear resistance [149]. In addition, investigations into Ta-C coatings designed with alternating hard and soft layers [150], as illustrated in Fig.7(c), have demonstrated that this configuration not only effectively reduces friction and wear under conventional conditions but also offers a unique advantage in regulating tribological performance at elevated temperatures. The key to this advantage lies in the ability of the alternating soft-hard design to modulate the structural stability and friction behavior of the coating at high temperatures. The soft layer, with its toughness and cushioning effect, helps to reduce brittle fracture at high temperatures, while the hard layer preserves the coating's high hardness and wear resistance, thereby achieving superior tribological performance in extreme operating conditions. These findings provide essential theoretical and experimental support for the design of alternating soft and hard structures in DLC coatings, further advancing the optimization and application of their tribological properties.

In addition to the alternating multilayer structural design intrinsic to DLC itself, numerous scholars have devoted considerable efforts to the strategic design of alternating composites between DLC coatings and other functional materials, achieving a series of groundbreaking advancements in this domain [143, 151-153]. For instance, to achieve ultra-lubricating performance under ultra-high contact stresses, some researchers have developed multilayer coatings reinforced with Si-DLC/PLC nanostructures [151]. Through the implementation of reciprocating friction tests with alternating loads, it was ascertained that this Si-DLC/PLC nanostructured multilayer coating exhibited unparalleled load adaptability at the sliding interface, as shown in Fig.7(d). Specifically, the coating underwent in-situ nano-agglomeration transitions during friction and localized ordering in the $\text{sp}^2\text{-C}$ region, thereby ensuring the maintenance of ultra-lubricity under alternating load conditions. The study emphasizes the pivotal role of the Si-DLC/PLC alternating structure, attributing its effectiveness to the

significantly relaxed internal residual stresses and the formation of newly grown nanostructures at the sliding interface. These features ensure the coating's stability and ultra-lubricity under high contact stresses and complex friction conditions. Concurrently, SiC-*n*WCC multilayer coatings have provided a benchmark for the exceptional lubrication performance of DLC coatings at elevated temperatures (see Fig.7(e)) [153]. The exceptional lubricity of SiC-*n*WCC multilayer coatings can be attributed to their remarkable thermal stability and the development of temperature-adaptive tribological layers. Furthermore, in response to stringent demands for low friction and extended service life in key moving components of spacecraft operating under alternating atmospheric and vacuum environments, researchers innovatively designed and developed an environment-adaptive LaF₃-Ti-MoS₂/Ti-DLC (MTLC) multilayer lubricating coating as shown in Fig.7(f). This coating strategically combines the excellent vacuum lubrication performance of LaF₃-Ti-MoS₂ with the low-friction superiority of Ti-DLC under atmospheric conditions. The precise regulation of the substrate's residence time in the proximity of each target during the magnetron sputtering process has been demonstrated to facilitate the in-situ self-assembly and precise construction of the multilayer architecture. The experimental findings indicate that when the substrate rotation speed was optimized to 0.5 rpm, the MTLC coating demonstrated not only exceptional mechanical properties but also a stable service life that exceeded 200,000 cycles under simulated alternating atmospheric-vacuum conditions. The findings indicate its potential for implementation and engineering viability in extreme aerospace environments [154].

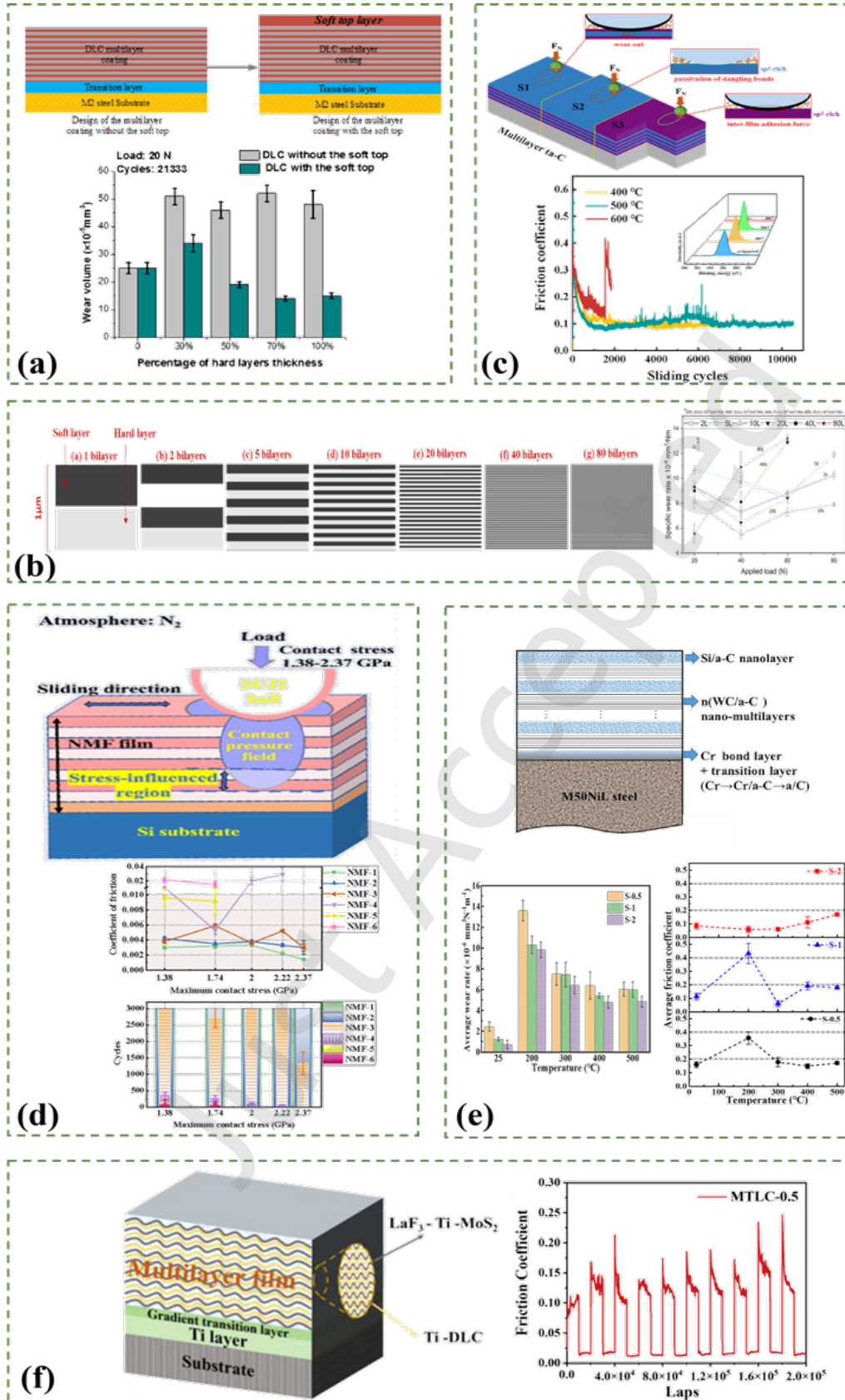


Fig. 7 (a) Wear volume of alternating soft and hard structural coatings [145]. (b) Wear rate of coatings with different alternating cycles [146]. (c) Friction coefficient of multilayer ta-C coatings [150]. (d)

Characterization of tribological properties and contact stress distribution of alternate coatings [151].

(e) Tribological performance characteristic of nanocomposite multilayer SiC-*n*WCC coating [153]. (f)

Tribological properties of LaF₃-Ti-MoS₂/Ti-DLC multilayer lubricating coatings [154].

Collectively, the alternating structural design of DLC coatings exhibits outstanding performance under extreme service conditions, including high temperatures, high pressures, and alternating friction. These advantages stem from the rational arrangement of material layers, which offers a novel conceptual framework and valuable reference for the development and engineering application of advanced high-performance coating technologies.

3.1.2 Gradient structure

While alternating structure design enhances the localized properties of DLC coatings, multilayer gradient structure design provides a more continuous and progressive approach to property modulation [155-157]. In contrast to the abrupt transitions in material properties inherent to alternating architectures, the gradient structure realizes a smooth transition of physical and chemical properties from the substrate to the surface by introducing gradient evolution of hardness, microstructure, or chemical composition along the thickness of the coating [59, 158-162]. This gradient functionalization design concept emphasizes the structural continuity and performance coordination from the inside out. This gradient functionalization concept emphasizes structural continuity and coordinated performance throughout the entire depth of the coating. By fundamentally mitigating interfacial mismatches, it facilitates the synergistic integration of multiple functional regions, thus imparting the coating with more stable, reliable, and controllable comprehensive performance [163, 164].

The gradient architecture of the material under consideration provides an optimized balance among hardness, toughness, wear resistance, and interfacial adhesion. This is accomplished through an effective mitigation of the intrinsic contradictions arising from complex stress fields and demanding frictional conditions. Consequently, the coating demonstrates augmented performance in multifaceted mechanical and tribological environments, owing to its superior comprehensive adaptive capacity. It

has been demonstrated that the gradient structural design has the capacity to significantly alleviate residual stresses within the coating, thereby strengthening the interfacial bonding between the coating and the substrate. This enhancement in interfacial integrity contributes to a marked improvement in both the structural stability and the operational longevity of the coating. For instance, several studies have demonstrated that the CrN/W-DLC/DLC gradient structure effectively reduces internal stresses, while simultaneously improving the wear resistance of the coating, thereby extending its service life [165]. Moreover, the gradient design optimizes the coating's load-bearing capacity and hardness by adjusting the coating thickness, enabling it to exhibit superior wear resistance even under harsh conditions such as high loads and friction [165]. In terms of specific applications, Ti/TiN/DLC gradient coatings [166] and CrN/TiN gradient coatings [167] have demonstrated remarkable improvements in the wear resistance and rolling contact fatigue life of bearing steel substrates. The gradient Ti/TiN/Si/(TiC/a-C:H) multilayer coatings enhanced the load-bearing capacity of the coatings by microscopically controlling the composition and structure of the sub-layers, while simultaneously reducing internal stresses to a significant degree [168]. On the other hand, gradient coatings fabricated via nitrogen ion implantation further improved the friction and wear performance by increasing the interfacial strength between the substrate and the coating, as well as enhancing the adhesion between the individual layers [169]. In gradient coatings composed of W-based carbides and nitrides, the transition layer structure provides exceptional tribological performance under heavy loads, owing to its ability to reduce the contact depth between the friction counterparts and increase the overall hardness of the coating structure [59, 161].

Guided by the design concept of functionalized gradient multilayer architectures, a sophisticated gradient coating system was fabricated via the closed-field unbalanced magnetron sputtering technique [170], comprising a Ti primer layer, a Ti/C gradient transition layer, a Ti-C composite layer, and a DLC top layer. When the Ti target current was set to 0.4 A, the coating achieved an impressive hardness of 44.6 GPa, representing a high-performance benchmark among current reports. At a

reduced Ti target current of 0.3 A, the coating exhibited an ultralow coefficient of friction (0.02), along with excellent resistance to both wear and corrosion. These results highlight the synergistic interaction between nano-TiC particles and the amorphous carbon matrix, while affirming the critical role of gradient structuring in modulating coating performance. Furthermore, the coating maintained its amorphous structure even after high-temperature annealing at 600 °C, and only at higher temperatures did the precipitation of graphitic carbon and TiC nanocrystals occur, which demonstrates its structural stability and suitability for demanding high-temperature engineering applications [171]. In a related study, a multilayer gradient architecture consisting of Ta, TaN, Ta(C, N), and Ta-DLC was fabricated using high-power pulsed magnetron sputtering [172], where modulation of the periodic structure enabled simultaneous enhancement of mechanical and tribological properties. At an optimal modulation period of 305 nm, the coating exhibited a hardness of 24.5 GPa, a reduced friction coefficient of 0.15, and a notably low wear rate of $2.47 \times 10^{-12} \text{ m}^3/\text{N} \cdot \text{m}$. The incorporation of the Ta-doped gradient structure led to a substantial enhancement in interfacial adhesion and overall performance, thereby demonstrating the distinct advantages of gradient multilayer engineering in the development of high-performance coatings. Additionally, another study presented a Cr/Cr-C composite interlayer between the DLC layer and substrate to further enhance interfacial bonding. a-C:Ta composite coatings were deposited via physical vapor deposition, with the rotational speed of the substrate carefully controlled. It was found that increasing the rotational speed led to a gradual transition in coating morphology from a dense amorphous structure to a columnar growth mode, accompanied by an increase in sp^2 -hybridized carbon content and a decrease in the proportion of C-Ta bonds [173]. This morphological evolution enhanced interfacial adhesion, although it resulted in a decline in wear resistance. Tribological testing revealed that all samples exhibited stable frictional behavior, with the lowest wear rate observed at 0.5 rpm, which is attributed to the reinforcing effect of the dense amorphous matrix and uniformly distributed TaC nanoclusters.

In the advanced development of multilayer DLC architectures, researchers have constructed a Cr/CrC/DLC gradient multilayer coating system, as shown in Fig.8(a). This coating demonstrates noteworthy tribological performance across a substantial temperature range when applied to a 17-4PH stainless steel substrate [174]. The experimental findings suggest that within the temperature range of 25 to 200°C, the coating attains an ultra-low coefficient of friction, with values as low as 0.03, while concurrently exhibiting exceptional wear resistance. These findings underscore the material's exceptional lubricating properties under variable thermal conditions. Subsequent mechanistic investigations have elucidated that the incorporation of Cr/CrC transitional layers substantially augments the interfacial adhesion strength between the coating and the metallic substrate. During the friction process, these layers work in conjunction with Si₃N₄ counterparts to collectively impede the graphitization of the DLC layer under high-temperature loading and concurrently promote the formation of a stable lubricating transfer film. This dual mechanism effectively delays the deterioration of wear behavior from abrasive wear to adhesive wear and delamination failure, thereby substantially extending the service life of the coating. Meanwhile, in the realm of gradient interlayer configuration design and plasma-assisted interfacial engineering, researchers have proposed pragmatic and efficacious strategies to address the adhesion challenges of a-C:H coatings on steel substrates, as illustrated in Fig.8(b). In this context, a gradient a-C:H/a-SiC_x:H transitional coating with excellent adhesion was successfully fabricated at relatively low deposition temperatures using tetramethylsilane as the precursor [175]. Interfacial chemical analyses demonstrate that the gradient interlayer formed under a low-oxygen atmosphere effectively suppresses oxygen incorporation at the interface, thereby significantly enhancing the bonding strength between the coating and the substrate. Subsequent interface modulation was achieved through the exposure of the silicon-containing interlayer to nitrogen plasma. Despite the interlayer thickness remaining relatively constant, a significant reconstruction of chemical bonds was observed within the interfacial region. It is noteworthy that after a duration of 20 minutes of plasma treatment, the interface demonstrated a high nitrogen-to-oxygen ratio and a low

silicon-to-carbon ratio. This chemical environment effectively promoted interfacial bonding strength. Consequently, a substantial enhancement in the adhesion performance of the a-C:H coating was achieved [176]. These findings demonstrate that by precisely tuning gradient structural design and interfacial chemical architecture, it is possible to optimize interfacial stability and overall tribological performance simultaneously. This, in turn, establishes a solid foundation for the reliable application of DLC coatings under complex service conditions.

In summary, the multilayer structure design not only optimizes coating performance from various aspects (see Fig.8(c)), such as enhancing support, reducing contact, dispersing stress, and chemical regulation, demonstrating its unique advantages, but also significantly improves the environmental adaptability and long-term stability of the coatings. This design concept offers valuable insights for the innovation and development of coating technology, establishing a robust foundation for its reliable long-term application under complex operating conditions. Consequently, it facilitates the widespread adoption of coating technology across various engineering fields.

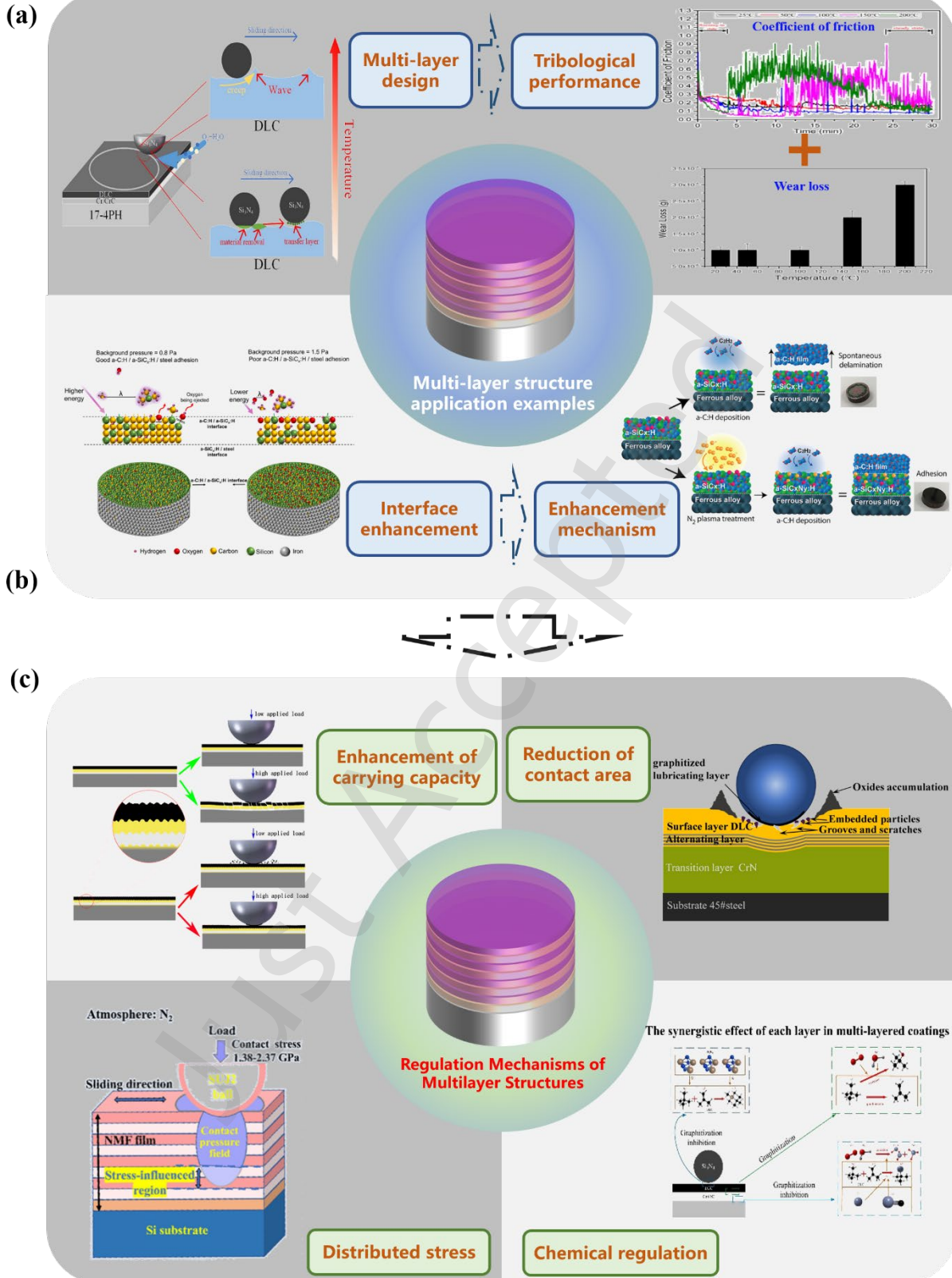


Fig. 8 (a) Schematic representation of friction mechanism, tribochemical reaction, coefficient of friction and wear loss [174]. (b) Interface engineering mechanism of a-SiC_x:H interlayer for enhancing

coating adhesion performance [175, 176]. (c) Regulatory mechanism of multilayer structure design of DLC coatings [151, 165, 169, 174].

3.2 Heterogeneous element doping

Heterogeneous element doping is an effective strategy for improving the mechanical properties, tribological behavior, and environmental adaptability of DLC coatings [177-182]. According to the different modification mechanisms and functions of dopant elements, current studies mainly focus on three categories: metallic element doping, non-metallic element doping, and multi-element co-doping strategies [81, 183-189]. In this section, recent advances in these three doping approaches are systematically reviewed to provide guidance for the design of high-performance DLC coatings under extreme aerospace environments.

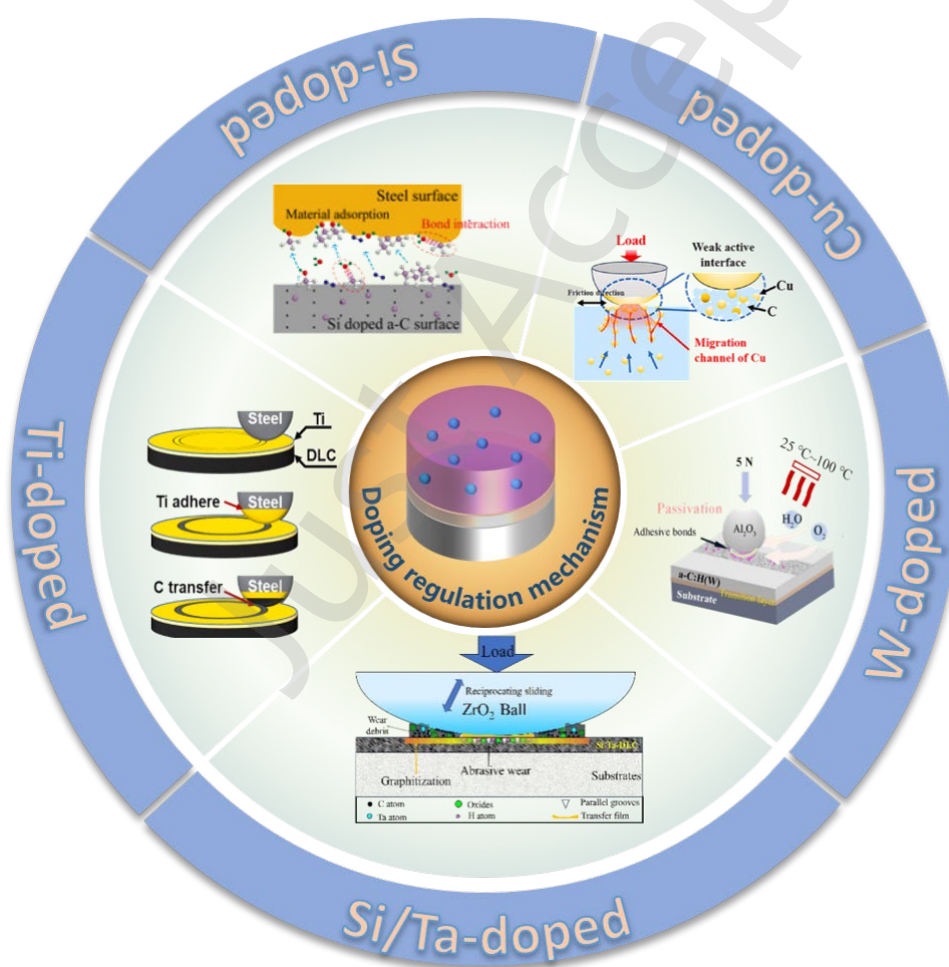


Fig. 9 Modulation mechanism of doping with different heterogeneous elements in DLC coatings [190-194].

3.2.1 Metallic element doping

Metal elements are commonly incorporated into DLC coatings in the form of atomic-scale solid solutions, nanocrystalline dispersions, or metal carbide phases [81, 180, 195, 196]. These constituents are typically non-uniformly distributed and highly dispersed within the amorphous carbon-based three-dimensional network, giving rise to composite nanostructures that manifest synergistic functionalities across multiple phases and hierarchical length scales [197, 198]. The introduction of such new phases contributes to significant enhancements in mechanical toughness, thermal stability, and tribological performance, including improved friction reduction and wear resistance, thereby exemplifying the multifunctional benefits of the “one dopant, multiple effects” strategy [168, 189, 197, 199]. The structural evolution and property modulation mechanisms induced by metal doping are profoundly influenced by the specific dopant species, rendering the effects of doping highly element-sensitive.

The doping of trace metal elements has been demonstrated to effectively alleviate the intrinsic residual stress within DLC coatings while significantly enhancing the adhesion between the coatings and their substrates [200]. However, the mechanisms through which different metal dopants modulate the properties of DLC coatings exhibit substantial variation and are highly dependent on the intrinsic nature of the dopant species [197]. Representative metallic dopants such as Cr [61, 80, 200-206], Ti [75, 190, 207-211], and W [47, 63, 116-118, 191, 212-215], owing to their high chemical reactivity, readily interact with carbon atoms within the DLC matrix to form MeC-type metal carbide nanophases [197, 207]. The formation of these carbides not only alters the microstructural configuration of the coatings, but also induces a transformation of sp^3 -hybridized carbon into sp^2 -hybridized carbon, thereby exerting profound influences on the coatings' hardness, density, and wear resistance [190, 201, 204, 212]. It is particularly noteworthy that the incorporation of metal carbides often leads to a marked reduction in coating brittleness, accompanied by enhancements in toughness and crack resistance, which collectively contribute to improved structural stability and reliability of the coatings under high-stress and extreme environmental conditions [54, 189, 200, 214]. Moreover, the doping concentration

of metal elements plays a pivotal role in dictating the comprehensive performance of DLC coatings [200, 202, 204, 211, 213]. Beyond these performance enhancements, the introduction of active metal dopants further endows DLC coatings with a suite of functional improvements. For instance, as depicted in Fig.10(a), investigations on Ti-doped DLC coatings have revealed that the Ti transfer layer serves as an adhesive intermediary between Fe and C atoms, facilitating the formation of stable carbon transfer layers and enabling low-friction behavior. Ti also exhibits strong adhesion to steel balls, forming durable and stable carbon transfer layers during operation, a mechanism that significantly bolsters the coatings' durability [190]. Similarly, as illustrated in Fig.10(b), W-doped DLC coatings demonstrate exceptional performance in oxygen-rich environments, owing to the reactivity of the W atoms with adsorbed oxygen, resulting in the formation of W oxides that possess remarkable friction-reducing properties [116-118, 191]. Less common, yet equally noteworthy, is the doping of reactive metals like Mo, which has also been found to enhance DLC coating performance. Research on Mo-doped DLC coatings has demonstrated a substantial increase in the coatings' critical failure load, attributed to improved ductility and metal-induced graphitization of the amorphous carbon matrix, which reduces residual compressive stresses [62, 216-218].

Besides these, according to Fig.10(c), the uniform incorporation of trace amounts of amorphous Ti into the DLC matrix has been shown to significantly enhance the wear resistance of the resulting coatings under dry sliding conditions, achieving an exceptionally low wear rate of $3.13 \times 10^{-11} \text{ mm}^3/(\text{N} \cdot \text{mm})$ after one million sliding cycles. This remarkable improvement is primarily attributed to the ultrafine scale and homogeneous distribution of amorphous Ti nanoclusters, which promote the formation of atomically smooth surface structures, facilitate the ordered alignment of sp^2 -hybridized carbon domains, and effectively relieve internal residual stresses within the coating [75]. Through precise regulation of the Ar/CH₄ gas flow ratio, low-concentration Ti doping can be achieved without inducing the formation of TiC phases. It has been demonstrated that even trace levels of Ti, when introduced under these controlled conditions, can markedly mitigate internal stresses while preserving

the intrinsic high hardness of the DLC matrix. Notably, this doping approach reduced the coefficient of friction to an ultralow level of 0.05 and resulted in a substantial reduction in wear rate, approaching a decrease of nearly two orders of magnitude [207]. Therefore, the fine-tuned control of Ti doping state and concentration, which enables a moderate degree of structural softening while maintaining the structural integrity of the carbon-based framework, represents a critical pathway toward the synergistic optimization of DLC coating properties.

In the amorphous carbon matrix of diamond-like carbon DLC coatings, metal elements with high chemical inertness, characterized by a weak bonding tendency with carbon atoms, typically exist in two structural forms: as atomic-scale solid solutions or as nanoscale dispersed phases [197]. Due to their low chemical affinity for carbon, these metal elements rarely form stable covalent bonds. Instead, they are commonly incorporated into the carbon network as isolated metallic nanoparticles or atom clusters. This distinctive microstructural configuration exerts a considerable influence on the macroscopic physical properties of DLC coatings, particularly in modulating their mechanical characteristics, including hardness, fracture toughness, and tribological behavior. For instance, studies on Al-doped ta-C coatings have demonstrated that, at low doping concentrations, the coating preserves its amorphous structure with aluminum atoms uniformly dispersed within the tetrahedral carbon network in a solid-solution state. As the doping concentration increases, aluminum begins to precipitate as discrete nanoparticles that become embedded within the carbon matrix. At even higher concentrations, the carbon structure undergoes a transformation toward graphitization, induced by the nucleation and growth of nanoscale aluminum particles. This structural evolution, resulting from the combined effects of stress relaxation introduced by aluminum incorporation and the progressive graphitization of the carbon matrix, leads to a significant reduction in residual stress, though accompanied by a noticeable decrease in hardness and elastic modulus. More importantly, Al doping substantially enhances the tribological performance of the DLC coatings, as evidenced by improved wear resistance at room temperature and, more strikingly, by outstanding frictional behavior at

elevated temperatures up to 400 °C, where the coating achieves a low coefficient of friction of 0.29 and an ultralow wear rate of $12.3 \times 10^{-8} \text{ mm}^3/\text{N} \cdot \text{m}$, thereby demonstrating exceptional anti-friction and anti-wear capabilities under high-temperature service conditions [219-222].

Moreover, studies on the doping of noble metals such as Cu [192, 223-227], Ag [228-232], and Au [233-235] have demonstrated that, due to their inherent difficulty in forming stable chemical bonds with carbon atoms, these metals are predominantly embedded within the amorphous carbon matrix as either amorphous phases or nanocrystalline clusters, thereby constituting a characteristic nanocomposite structure. The effect of metallic dopants on the bonding configurations of C-C bonds within the coating is not significant. However, through lattice filling and interfacial reinforcement effects, metallic dopants can effectively modulate the internal stress and brittleness of the film. During frictional processes, the high chemical reactivity of noble metal nanoparticles facilitates their migration toward the interface under the synergistic influence of frictional heating and shear forces. This results in the formation of metal-rich transfer films with intrinsic self-lubricating properties, which, in concert with the graphite-rich transfer layers induced by friction, contribute to a notable reduction in the coefficient of friction and an enhancement in the wear resistance of the DLC coatings [223, 228, 233]. For instance, as reported in the extant literature and illustrated in Fig.10(d), during tribological testing under vacuum conditions, the interaction between carbon and copper atoms is weaker than that of C-C bonds. Consequently, under the combined effects of frictional heat and mechanical load, Cu atoms tend to migrate within the coating and gradually coalesce into larger crystalline clusters. These clusters can be transferred along the migration pathways to the tribological interface, eventually forming a Cu-rich transfer film on the counterface steel ball. This transformation effectively converts the originally reactive a-C:H/C interface into a more tribologically stable Cu/C interface, thereby substantially extending the service life of Cu/a-C:H coatings under vacuum conditions [192]. Another study prepared Ag-DLC coatings with Ag contents ranging from 1.3 to 13.1 at.% via DC magnetron sputtering and systematically investigated the influence of Ag doping on the tribological performance

of the coatings. The study found that when the Ag content was below 6.1 at.%, Ag existed mainly as highly dispersed nanoclusters, exerting minimal impact on the carbon-based structure and mechanical properties of the DLC. However, when the Ag content increased to 13.1 at.%, crystalline Ag particles with sizes of approximately 2-3 nm formed within the coating, promoting graphitization and leading to a significant decrease in hardness and compressive residual stress. Tribological tests indicated that the effect of Ag doping was highly dependent on the contact pressure. Under lower contact pressure (690 MPa), the wear rates of the coatings showed only minor changes; however, when the contact pressure was increased to 1180 MPa, the wear rate of the Ag-DLC coatings was significantly higher than that of the pure DLC coatings. SEM analysis further revealed that during sliding, Ag segregated and accumulated along the wear tracks and on the counterface, forming an Ag-rich transfer layer. This Ag-rich layer gradually replaced the original low-shear carbon-based transfer film, enhancing interfacial adhesion and intensifying abrasive wear, ultimately leading to significantly increased friction coefficients and wear rates [229]. Additionally, a study reported a unique interaction between Au nanoparticles and carbon dangling bonds within the amorphous carbon network, based on which a “self-migrating intelligent lubrication” strategy was proposed to address the lubrication failure of conventional hydrogen-free DLC coatings under vacuum conditions. This interaction enables Au atoms to remain stably dispersed within the a-C network and gradually self-migrate toward the counterface during sliding, ultimately forming an Au-C tribological interface with excellent lubricating properties. Tribological results demonstrated that the film achieved an ultralow friction coefficient below 0.03 under vacuum conditions and maintained extremely low wear after 6.0×10^5 sliding cycles, exhibiting outstanding lubrication lifetime and service stability. Combined experimental analyses and molecular dynamics simulations revealed that the superior tribological performance mainly originated from the formation of a low-shear and self-repairing lubricating interface composed of Au-rich and carbon-rich layers generated during sliding, which enabled the interface to continuously maintain stable lubrication even after local damage [234].

On the whole, the incorporation of trace metal elements can significantly enhance the overall performance of DLC coatings, with the specific modulation mechanisms largely dependent on the nature of the dopant. Highly reactive metals such as Cr, Ti, and W readily form carbide nanocrystalline phases with carbon atoms within the DLC matrix. This interaction facilitates the transformation of sp^3 -hybridized carbon into a more unsaturated sp^2 configuration, while concurrently relieving internal stresses and enhancing the coating's toughness, lubricity, and wear resistance. In contrast, chemically inert metals such as Al, Cu, Ag, and Au are typically incorporated into the amorphous carbon network in the form of nanocrystals or atomic-scale solid solutions. Rather than altering the carbon bonding configuration directly, these elements modulate the mechanical and tribological properties of the coating by adjusting its microstructure and electronic band structure. Additionally, variations in doping concentration exert a considerable influence on the structural organization and interfacial reactivity. For DLC coatings designed for aerospace applications under extreme service conditions, the rational selection and precise control of both dopant type and content are therefore essential to achieving high-performance and durable coatings.

3.2.2 Non-metallic element doping

The introduction of non-metallic elements into DLC coatings generally occurs through the substitution of a portion of sp^3 -hybridized carbon atoms or hydrogen atoms, resulting in the formation of new sp^3 bonds with neighboring carbon atoms [193, 236]. This modification regulates the ratio of hybridized carbon within the coating, thereby inducing a reorganization of the carbon-based network structure and ultimately enhancing the overall performance of the coating [182, 237-239]. A notable example is Si doping, which has been demonstrated to enhance coating performance through various mechanisms. As illustrated in Fig.10(e), Si doping has been shown to enhance H-H interactions through the process of terminal passivation of surface dangling bonds with hydroxyl (-OH) or hydrogen (-H) groups. Secondly, it has been demonstrated that this process induces $\pi^*-\pi^*$ interactions between graphitized layers, thereby significantly reducing the friction of the coatings [193]. An

alternative perspective posits that the graphite and O-Si-C structures present within the transfer layer can effectively diminish shear resistance and function as lubricants during the friction process [240]. Si doping is a pivotal strategy for surmounting the wear resistance limitations of DLC coatings under elevated temperature operating conditions. A multitude of studies have demonstrated that Si-DLC coatings demonstrate exceptional thermal stability and remarkably low coefficients of friction at elevated temperatures ranging from 400 °C to 500 °C [177, 241-244]. This enhancement in performance is primarily attributed to the fact that silicon doping suppresses the tendency for graphitization at elevated temperatures, while also providing additional lubrication through the formation of Si-O-C bonds during sliding [177, 243]. With regard to F doping, some researchers have found that F-DLC coatings exhibit extremely low coefficients of friction in humid environments. This can be explained by the higher interfacial force required for fluorine to bind with -OH compared to -H, thereby imparting superior hydrophobicity and friction-reducing properties to F-DLC coatings [177, 245].

In addition, investigations into the tribological behavior of N-DLC coatings have revealed that the applied bias voltage plays a pivotal role in modulating both the microstructure and performance of the coatings [245-248]. As demonstrated in Fig.10(f), increasing the bias voltage facilitates the incorporation of softer secondary phases, such as (CN)_x species and graphite-like structures, into the amorphous carbon matrix. While the presence of these phases may compromise the coating's hardness and mechanical robustness to some extent, they are highly susceptible to friction-induced graphitization under the influence of interfacial thermal effects. This transformation promotes the formation of a lubricious, graphitic-rich transfer film at the sliding interface, which exhibits remarkable self-lubricating characteristics and significantly enhances the overall tribological performance of the coating [246].

In summary, non-metallic element doping is a vital approach for regulating the performance of DLC coatings. Different dopants exhibit distinct bonding behaviors and structural evolution trends within

the carbon matrix. For example, silicon tends to form stable Si-C bonds, fluorine imparts low surface energy to the coating, and nitrogen facilitates the incorporation of (CN)_x groups or promotes the formation of graphite-like phases. These varied bonding characteristics directly influence the mechanisms of transfer film formation and interfacial reactions during friction. Si-doped DLC coatings achieve wear resistance through the formation of stable silicon oxide transfer films. F-doped DLC coatings reduce adhesive friction by weakening interfacial interactions. N-doped DLC coatings develop graphitized transfer films that provide adaptive lubrication. This clear correlation between elemental doping, structural transformation, and tribological behavior offers a theoretical basis for the functional design and engineering application of non-metal-doped DLC coatings in extreme service environments.

3.2.3 Multi-element co-doping

Multi-element co-doping of DLC coatings aims to integrate the performance advantages of various dopant species and achieve synergistic regulation of the coating's structure and properties at the atomic scale [194, 244, 249-251]. This strategy overcomes the inherent functional limitations of single-element doping and fosters the integration of multiple performance attributes within the coating, thereby advancing its applicability in complex and demanding service environments [252]. For instance, in studies investigating the synergistic modulation of high-temperature tribological properties of DLC coatings by Si and W, it was revealed that the cooperative effects of coating graphitization, the formation of WO₃, WO₂, WC, and silicon oxides on the wear track, coupled with the mechanical property improvements brought about by the gradual distribution of W elements, are crucial factors contributing to the superior tribological performance at elevated temperatures [153]. It is widely acknowledged that DLC coatings are susceptible to graphitization and severe oxidation, which can lead to failure under high-temperature conditions. However, the co-doping of Ta and Si led to the formation of Ta₅Si₃ binary compounds and SiC nanoparticles, as illustrated in Fig.10(g). This dual doping not only preserved the structural integrity of the coatings to a considerable extent but also

significantly enhanced their tribological performance [194]. The W and Mo co-doped DLC coatings form a transfer film during the friction process, which is enriched with amorphous graphite structures containing substantial amounts of W and Mo carbides and oxides, thereby exhibiting exceptional lubrication performance [253]. In studies on F and Si co-doped H-DLC coatings, it was observed that the introduction of Si increased the hydrogen atom content in the coatings, which indirectly led to a decline in the mechanical properties of the coatings, yet yielded a lower coefficient of friction. Conversely, the addition of F improved the hardness and wear resistance of the coatings, with the combined doping of Si and F providing a highly complementary effect [254].

The aforementioned studies demonstrate that the multi-element co-doping strategy for DLC coatings constructs a structure-function correlation mechanism through the coordinated evolution of elemental combinations, bonding states, and performance outcomes. This approach facilitates precise modulation of multiple functional properties at the atomic scale. It provides important insights into how elemental doping influences the microstructure and interfacial behavior of DLC coatings, and offers valuable theoretical and practical guidance for the development of high-performance coatings suitable for extreme service conditions.

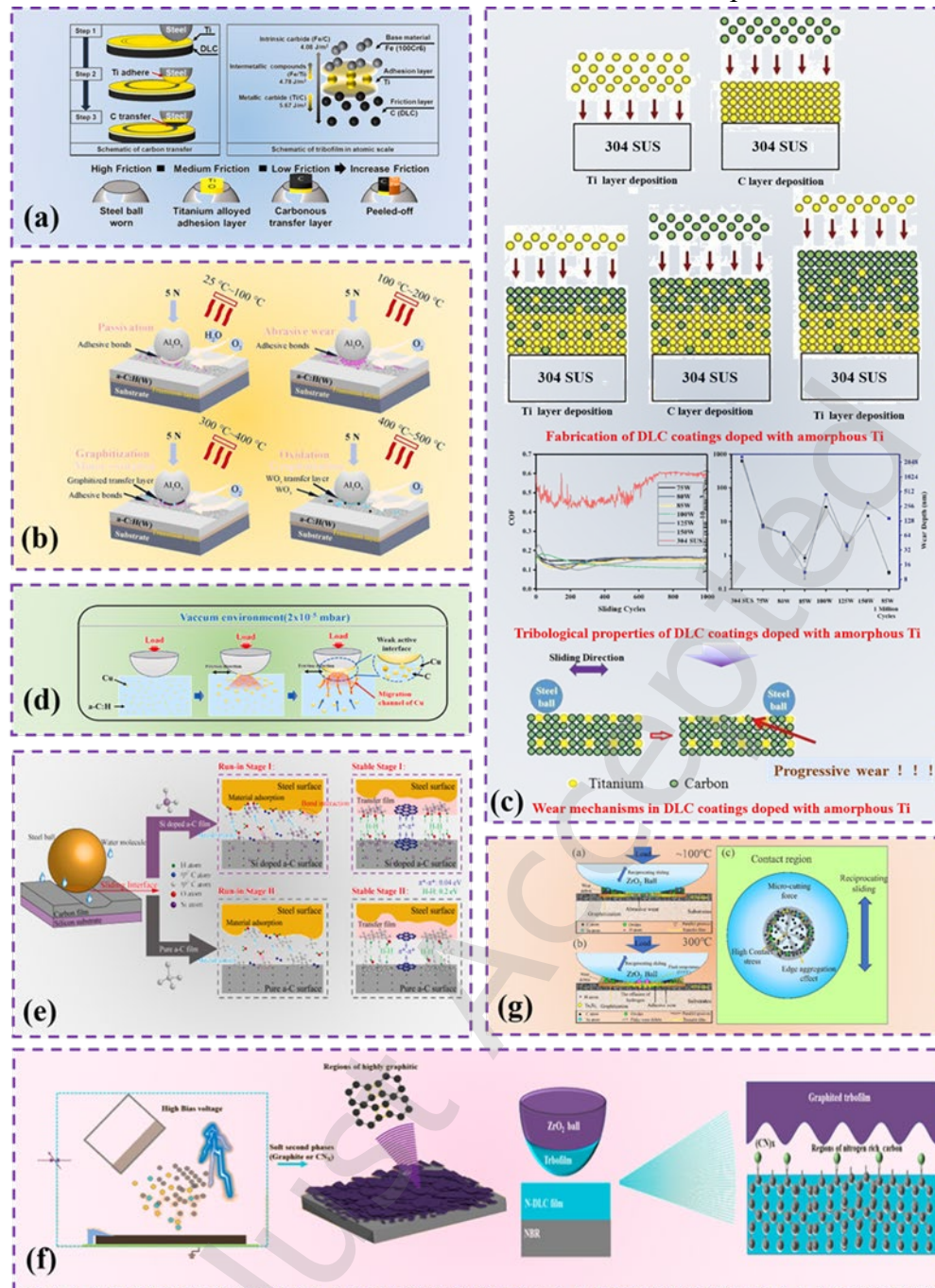


Fig. 10 (a) Suggested schematic of process of carbon transfer in Ti/DLC and tribofilm structure in atomic scale [190]. (b) schematic representation of the friction mechanism of a-C:H(W) coatings at different temperature [191]. (c) Preparation, tribological performance, and wear mechanisms of DLC coatings doped with amorphous Ti [75]. (d) Schematic diagram of the mechanism of anti-wear life extension of Cu/a-C:H films in vacuum [192]. (e) Low-friction mechanism of Si-doped a-C coating

[193]. (f) Preparation and friction reduction mechanism of N-DLC coating [246]. (g) Schematic diagram of related wear mechanism of Si/Ta-DLC films at different temperatures [194].

3.3 Surface morphology optimization

Conventional tribological theory generally assumes that the smoother the contact surface is, the lower the coefficient of friction and wear rate will be. However, numerous researches has demonstrated that the relationship between surface morphology complexity and friction behavior is significantly more intricate and nuanced than this traditional perspective implies [68, 255-257]. Therefore, optimizing the surface morphology of coatings to achieve superior tribological performance has become a significant direction of current research, particularly for DLC coatings, which exhibit high sensitivity to environmental factors [69, 257]. The optimization of surface morphology can enhance the overall performance of the coating across multiple dimensions, thereby significantly improving its tribological performance under extreme operating conditions. In this section, the research advancements in both surface roughness control and surface texture design of DLC coatings will be explored, with an analysis of how the tribological performance of DLC coatings can be further optimized through the precise regulation of their surface morphology.

3.3.1 Surface roughness control

Surface roughness plays a critical role in determining the tribological performance of DLC coatings. Variations in roughness directly influence the real contact area, local stress distribution, interfacial adhesion, and frictional behavior between the coating and its counterpart. More importantly, unlike conventional metallic or ceramic materials, the influence of surface roughness in DLC coatings is closely coupled with friction-induced graphitization, transfer film evolution, tribochemical reactions, and the dynamic reconstruction of the carbon bonding structure. Consequently, even slight variations in surface roughness can lead to significant differences in friction coefficient, wear resistance, and coating durability under complex operating conditions [258].

As illustrated in Fig.11(a), classical contact theories suggest that rough surfaces reduce the number of real contact asperities and increase localized contact stresses. In DLC coatings, however, this effect is uniquely manifested through its influence on transfer film formation and interfacial structural evolution. Excessive roughness may disrupt the continuity of carbonaceous transfer layers and induce localized stress concentration, thereby promoting microcrack propagation, abrasive wear, and local delamination of the coating [259-263]. In contrast, relatively smooth DLC surfaces are more favorable for the formation of continuous low-shear tribofilms, which effectively reduce interfacial adhesion and maintain low-friction sliding behavior.

Furthermore, due to the intrinsically high hardness and elastic modulus of DLC coatings, roughness-induced asperity interactions can generate pronounced local stress amplification, particularly in “soft substrate-hard coating” systems. This not only affects contact stiffness and load transfer behavior, but also alters frictional heat accumulation and tribochemical reaction processes at the sliding interface. For hydrogenated DLC coatings, surface roughness may additionally influence hydrogen passivation stability and surface adsorption behavior, whereas in doped DLC systems, roughness often interacts synergistically with oxide films or lubricating tribolayers induced by dopant elements.

As illustrated in Fig.11(b), numerical simulations based on deterministic adhesion models further reveal that surface roughness strongly regulates the adhesion behavior of DLC layered structures [264]. Compared with homogeneous materials, DLC/Si layered composites exhibit lower pull-off forces when contacting SiO₂ microspheres, while the adhesion attenuation induced by roughness becomes increasingly pronounced with increasing RMS roughness. However, once roughness exceeds a critical threshold, this influence gradually approaches saturation. These findings demonstrate that the coupled effect of surface roughness and interfacial layering is crucial for tailoring adhesion and tribological behavior in DLC coatings.

In the case of rough surface contact, tribological behavior is often dominated by mechanical interlocking, localized brittle fracture, and unstable transfer film evolution [265, 266]. Studies have

shown that the adhesion strength of rough DLC-coated surfaces and the microstructure of the transfer film play decisive roles in determining friction and wear performance [66]. Increasing coating fracture toughness can effectively suppress crack propagation and reduce the wear rate of rough DLC surfaces, thereby improving coating durability under severe operating conditions [266].

As illustrated in Fig.11(d), recent studies have demonstrated that surface functionalization using graphene sheets (GS) and nanodiamonds (ND) can effectively regulate surface roughness while promoting the formation of stable carbon-rich transfer layers during sliding [267]. The self-assembled tribofilm formed by GS, ND, and nanoscale carbonaceous structures exhibits excellent shear lubrication capability and structural stability, thereby significantly reducing friction coefficients and enabling robust ultra-low-friction performance under dry sliding conditions.

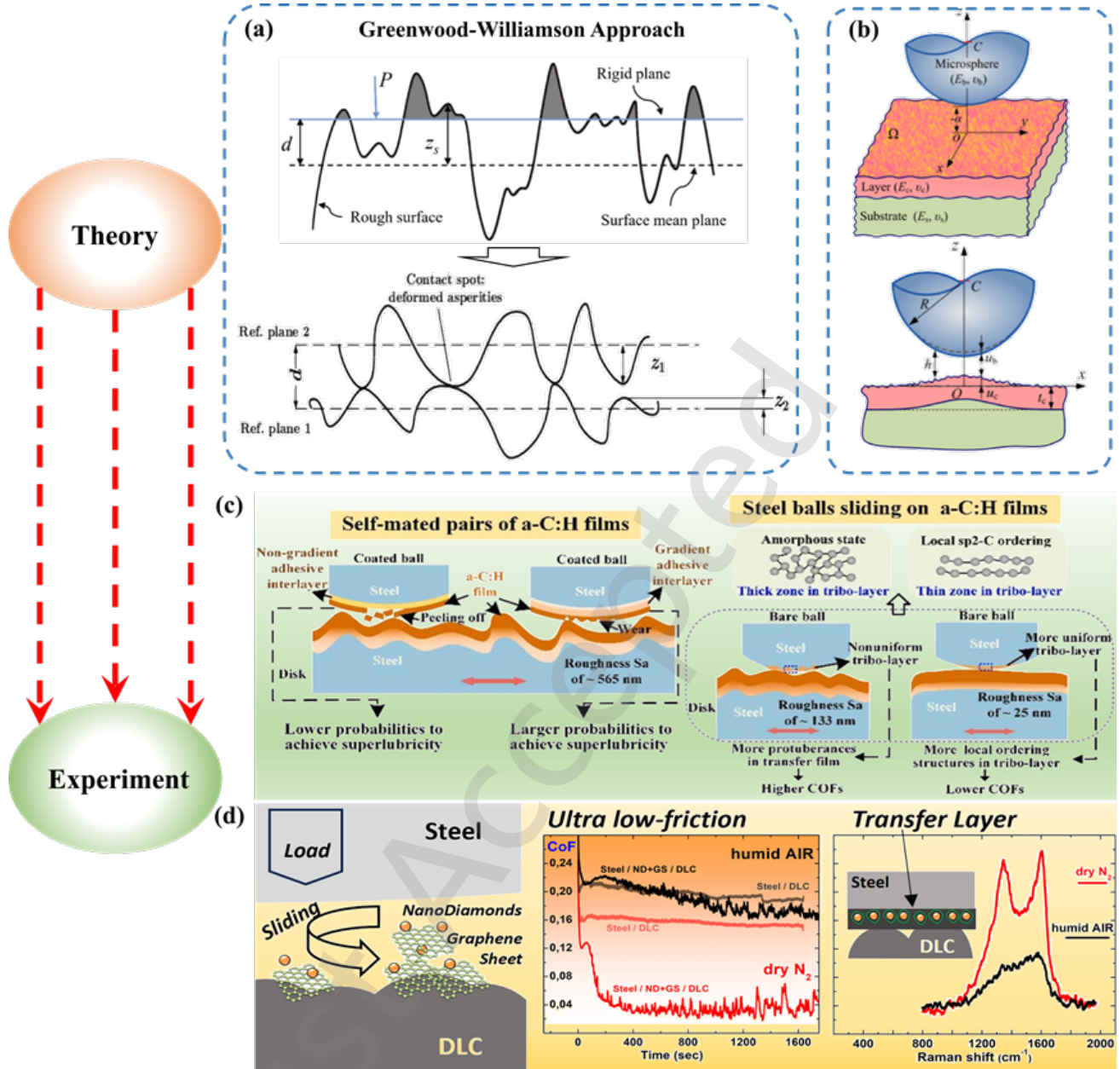


Fig. 11 (a) Graphical representation of the Greenwood-Williamson [260]. (b) Illustrations of a smooth elastic microsphere in adhesive contact with a rough-layered medium [264]. (c) Wear mechanism of DLC coatings on rough surfaces [66]. (d) Schematic illustration of the friction-reducing and wear-resistant mechanism of functionalized DLC coatings [267].

Overall, surface roughness control represents an effective strategy for optimizing the friction-reduction and anti-wear performance of DLC coatings. Nevertheless, because roughness effects are strongly coupled with coating structure, transfer film evolution, environmental atmosphere, and tribochemical reactions, further systematic investigations are still required to establish quantitative

relationships between roughness parameters and tribological performance under complex service conditions.

3.3.2 Surface texture design

Surface texture design, as illustrated in Fig.12(a), has emerged as an effective tribological optimization strategy that integrates structural functionalization with interfacial regulation. In recent years, it has attracted extensive attention in the field of solid lubrication owing to its capability to simultaneously improve friction reduction, wear resistance, and lubrication performance [268-274]. Through the deliberate construction of micro-/nano-scale surface features, textured surfaces can modify contact mechanics, regulate stress distribution, influence debris transport behavior, and alter lubricant flow dynamics. As a result, frictional losses can be effectively reduced while the service life of tribological components can be significantly extended [275]. When combined with DLC coatings, surface textures form a synergistic tribological system in which the superior hardness, chemical inertness, and low-friction characteristics of DLC coatings are coupled with the functional advantages of textured architectures [276, 277]. This combination is particularly beneficial under extreme operating conditions, such as vacuum, high temperature, oxygen-rich environments, and dry sliding conditions, where conventional lubrication mechanisms are often ineffective [67, 278].

The tribological performance of textured DLC coatings is strongly dependent on several geometric parameters, including texture shape, area density, depth, aspect ratio, and spatial arrangement [279-282]. Different texture geometries exhibit distinct tribological functions. For example, circular dimples are highly effective in trapping wear debris and storing lubricants, whereas groove textures facilitate directional lubricant transport and hydrodynamic pressure generation. Among these parameters, texture density is considered one of the most influential factors because it determines the balance between lubricant-storage capability and load-bearing area. Excessively low texture density limits debris-trapping and lubricant-retention capacities, whereas excessively high density reduces the effective load-bearing area and may compromise coating integrity. Previous studies suggest that

texture area densities ranging from 10% to 30% generally provide favorable tribological performance, although the optimum value varies with coating properties and operating conditions [279-282].

Representative studies have demonstrated the quantitative influence of texture geometry on tribological behavior. DLC surfaces with dimple densities ranging from 2% to 20% exhibited enhanced hydrophobicity, anti-adhesion behavior, and friction-reduction performance [69]. Adhesion tests revealed significantly reduced residue retention on textured surfaces compared with smooth DLC coatings. When the dimple density was optimized to approximately 10%, both friction and wear losses were substantially reduced, and the friction coefficient decreased by approximately 24% relative to that of a flat DLC surface. Furthermore, dimple textures exhibited superior wear resistance compared with groove-type textures, highlighting the critical role of texture geometry in determining tribological performance [69]. In addition to dimple structures, groove textures have also been extensively investigated under harsh operating conditions. Si/Ni-DLC and Si/Ta-DLC composite coatings deposited on textured SUS304 stainless-steel substrates exhibited improved tribological behavior over a temperature range of 25-300 °C [70]. Compared with untextured coatings, textured films showed lower wear rates while maintaining comparable friction coefficients at room temperature. The groove structures acted as reservoirs for wear debris and solid lubricants, thereby mitigating abrasive interactions and reducing stress concentration at the contact interface. Moreover, the textured architecture alleviated shear-stress accumulation around groove edges, suppressing crack initiation and propagation within the coating. At elevated temperatures, texture geometry exhibited a synergistic effect with coating composition. Nanocrystalline Si phases in Si/Ni-DLC coatings enhanced load-bearing capacity and wear resistance, whereas the combined action of TaC particles, Si, and oxygen in Si/Ta-DLC coatings promoted the formation of a compact Ta-Si-C-O tribolayer, thereby improving high-temperature tribological performance [70].

As illustrated in Fig.12(b), the tribological enhancement mechanisms of textured DLC coatings can be understood from multiple perspectives. First, textured features effectively capture wear debris

generated during sliding, preventing particle accumulation and repeated third-body abrasion at the contact interface, thereby reducing abrasive and adhesive wear [83, 283]. Second, the combination of DLC coatings and textured substrates forms a “rigid-flexible” coupling structure that enhances mechanical interlocking and suppresses ploughing-induced substrate deformation, resulting in improved wear resistance and load-carrying capacity. Third, the discrete contact characteristics introduced by texture units reduce the real contact area and redistribute local contact stresses, thereby decreasing interfacial adhesion and frictional resistance [83]. More importantly, in lubricated frictional systems, the pit-like textures serve as microscale lubricant reservoirs capable of adsorbing, temporarily storing, and subsequently releasing lubricant into the contact region. This dynamic replenishment mechanism mimics self-lubricating behavior, significantly retarding lubricant depletion and enhancing its utilization efficiency [83]. More importantly, in lubricated systems, textured structures act as microscale lubricant reservoirs capable of adsorbing, storing, and continuously releasing lubricant into the contact zone. This dynamic replenishment mechanism delays lubricant starvation and significantly improves lubricant utilization efficiency [83]. Simultaneously, optimized texture geometries can induce localized hydrodynamic pressure generation, increase lubricant film thickness, and improve film stability under varying loads and sliding speeds. Consequently, surface texture design not only reduces friction and wear but also enhances load-carrying capacity, lubrication efficiency, and operational reliability.

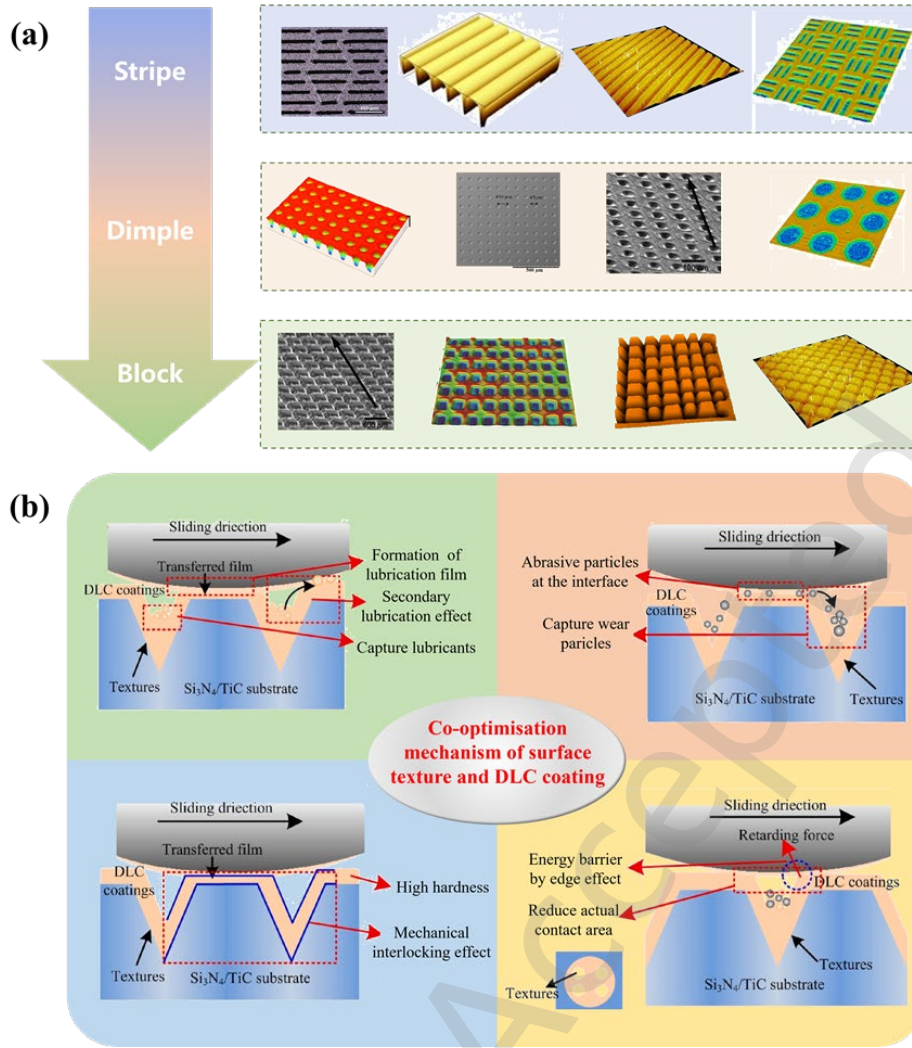


Fig. 12 (a) Typical surface morphologies of various geometrical texture configurations on the surface of the DLC coating [67-69, 276, 277, 279-282]. (b) schematic of synergistic effect mechanism of DLC coating and surface texture [83].

However, under dry friction conditions, the edge effect of surface textures becomes non-negligible. The reduction in nominal contact area may result in elevated localized contact stresses at the edges of the textured features, potentially inducing significant plastic deformation on the surface of the mating ball counterparts. Simultaneously, the micro-cutting action of the texture edges may facilitate the generation of additional wear debris, which tends to accumulate within the pits [48, 67]. Under high-temperature environments, the synergistic interaction between the surface texture and the DLC coating is primarily manifested through a combined mechanism involving the formation of a graphitization-induced transfer lubrication film, the effective entrapment of wear debris, and the reduction of actual

contact area [67, 284, 285]. These coupled effects collectively contribute to the enhanced friction-reducing and anti-wear performance of the coating system.

To conclude, surface texture design exhibits significant potential for enhancing the tribological performance of DLC coatings. Nevertheless, to further optimize texture design strategies and improve the stability and functional performance of coatings under complex and demanding service conditions, more comprehensive and in-depth investigations into the underlying mechanisms of surface texturing are still required.

In practical DLC coating design, multilayer structures, elemental doping, and surface morphology optimization are rarely considered in isolation. They need to be addressed in an integrated, system-level manner to achieve optimal performance in terms of mechanical properties, tribological behavior, wear resistance, and environmental durability. Multi-strategy synergistic design should focus on the interactions and potential coupling effects among different reinforcement approaches. For example, micro- and nanoscale surface textures can significantly enhance lubrication retention and wear resistance, but they may compromise the integrity of multilayer interfaces or induce local variations in dopant distribution, thereby affecting local hardness, residual stress distribution, and tribological performance. At the same time, different dopant elements contribute to DLC coatings through distinct mechanisms. Reactive metals such as W and Ti can strengthen the coating structure, whereas weak carbide-forming elements such as Al, Cu, and Ag mainly modulate frictional properties. With current atomic- or molecular-level deposition techniques (e.g., CVD, magnetron sputtering, ion beam-assisted deposition, and cathodic arc deposition), dopants are generally distributed uniformly, so macroscopic doping inhomogeneity is rarely observed [31, 47, 75, 116, 127, 220]. However, when multiple strategies are combined, texture scales are complex, or dopant concentrations are high, local stress concentrations, interface defects, or microstructural variations can still affect overall coating performance.

From a process-design perspective, the sequence of surface texturing and coating deposition is a critical factor for achieving synergistic optimization [282]. Two main approaches are commonly employed. The first, “texture first, coating later,” involves forming micro- or nanoscale textures on the substrate via laser processing, etching, or mechanical machining, followed by DLC deposition. This method ensures an integrated interface between the texture and the coating, enhancing adhesion, while allowing the coating to uniformly cover the textured surface and provide lubrication and debris-trapping functionality. However, complex textures may create local deposition shadowing, causing non-uniform coating thickness, and sharp edges can lead to stress concentration and potential cracking. The second approach, “coating first, texture later,” deposits a smooth DLC coating on the substrate before patterning the surface via laser or etching. This method can maintain uniform coating thickness and continuity while enabling precise control of texture dimensions, morphology, and distribution. Nevertheless, post-processing can induce local thermal damage, graphitization, residual stress changes, or interface weakening, which can reduce local load-bearing capacity and wear resistance.

Therefore, multi-strategy synergistic optimization requires not only consideration of material composition, interlayer structure, and dopant selection but also cross-scale control of macroscopic surface morphology, texture dimensions, and compatibility between deposition and processing techniques. By systematically analyzing the interactions, potential constraints, and synergistic effects among different reinforcement strategies, coating performance can be optimized at the system level, enhancing the overall functionality of DLC coatings under complex service conditions. This analysis elevates DLC coating design from the level of individual strategies to a system-level, multi-strategy coordinated approach, providing practical guidance and theoretical insight for future coating optimization.

3.4 Solid-liquid lubrication

Solid-liquid composite lubrication, as an advanced strategy for extreme service environments, has received increasing attention in recent years in aerospace, deep-space exploration, and cryogenic

mechanical systems. Diamond-like carbon (DLC) coatings, owing to their high hardness, low friction coefficient, excellent chemical inertness, and superior wear resistance, are considered highly promising solid lubricants for space applications. However, under complex operating conditions such as high vacuum, low temperatures, thermal cycling, and irradiation, DLC coatings still face challenges including enhanced adhesive wear, interfacial cold welding, and instability of transfer films. In particular, non-hydrogenated or low-hydrogen DLC can exhibit friction coefficients as high as 0.6-1.0 in ultra-high vacuum, severely limiting their engineering applicability. To overcome these issues, recent research has shifted from single-phase solid lubrication toward solid-liquid hybrid lubrication systems, in which liquid lubricants or functional additives are incorporated into DLC coating systems to construct interfaces with adaptive lubrication capabilities. The core of this strategy lies in utilizing DLC coatings to provide high load-bearing capacity and structural stability, while liquid lubricants form dynamically replenished and low-shear interfacial layers, thereby achieving multi-scale synergistic lubrication and friction regulation [286-289].

In terms of liquid lubricants, perfluoropolyether (PFPE), as a representative space lubricant, has been widely applied in DLC-based lubrication systems due to its low vapor pressure, excellent thermal stability, and chemical inertness. PFPE molecules possess a flexible ether-based backbone and highly fluorinated side groups, enabling them to maintain good fluidity and chemical stability over a wide temperature range [290-293]. When interacting with DLC surfaces, PFPE molecules can form a stable boundary lubrication film through physical adsorption and van der Waals interactions, effectively isolating direct solid-solid contact between friction pairs and significantly reducing interfacial shear strength, thereby achieving simultaneous reductions in both friction coefficient and wear rate [292]. In addition, during sliding, PFPE molecules can undergo local rearrangement and redistribution within the contact region, providing continuous lubrication replenishment to worn areas and exhibiting a certain degree of “quasi-self-healing” lubrication behavior [292]. However, PFPE-based systems still exhibit certain limitations in practical applications. Under long-term sliding or high-energy

environments (such as high vacuum, irradiation, or temperature fluctuations), PFPE molecules may suffer from volatilization loss, chain scission, or interfacial migration, leading to gradual thinning and eventual failure of the lubricating film. Moreover, since the interaction between PFPE and DLC surfaces is primarily governed by physical forces, the interfacial bonding strength is relatively weak, making it difficult to maintain stable lubrication under high load or boundary lubrication conditions for extended periods. In addition, in localized high-temperature friction zones, PFPE may undergo slight thermal degradation, further reducing its lubrication durability [290, 294].

In contrast, ionic liquids (ILs), due to their tunable molecular structures, near-zero vapor pressure, and excellent thermal stability, are considered a new generation of space lubricants [295]. IL molecules can form ordered adsorption layers on DLC surfaces through electrostatic interactions, hydrogen bonding, and van der Waals forces, thereby significantly reducing interfacial shear resistance and mitigating adhesive wear caused by direct solid-solid contact [296-298]. During sliding processes, the IL molecular layer can act as a dynamic boundary lubrication film, not only providing low-shear-strength channels in the contact zone but also adaptively regulating its molecular arrangement in response to variations in load, temperature, or sliding speed, thereby exhibiting a certain degree of “adaptive lubrication” behavior. More importantly, the lubrication performance of ILs strongly depends on their molecular structural characteristics. The type of cations and anions, alkyl chain length, polarity of substituents, and functional group distribution can all regulate their adsorption behavior on DLC surfaces, interfacial energy, and frictional response [297, 298]. For example, long-chain alkyl cations can form stable hydrophobic ordered layers on DLC surfaces, enhancing the thickness and uniformity of boundary lubrication films; ILs containing hydroxyl or carboxyl functional groups can form hydrogen bonds or weak chemical bonds with DLC surfaces, thereby further stabilizing the adsorption layer and improving wear resistance [299, 300]. In addition, the polarizability and charge distribution of different ion pairs can influence the lubrication behavior of ILs under high vacuum,

low-temperature, or high-temperature conditions, providing potential routes for friction regulation in extreme space environments [297, 298].

Recent studies have further demonstrated that ILs exhibit pronounced synergistic lubrication effects when combined with doped DLC coatings. Elemental doping with Ti, W, Eu, Gd, and others can significantly modify the surface chemical reactivity, bonding states, and electronic structure distribution of DLC coatings, thereby enhancing their interfacial compatibility with ionic liquids and promoting the formation and evolution of stable tribofilms [301-307]. During sliding, such interfacial films typically manifest as composite lubricating layers composed of adsorbed molecular films, tribochemical reaction products, and locally rearranged structures, which can effectively reduce interfacial shear strength and suppress adhesive wear and material transfer. For instance, W-doped DLC can enhance surface polarizability and active site density, thereby improving the adsorption capability and orientational ordering of ionic liquid molecules at the interface. This facilitates the formation of a stable tribochemical lubrication layer and significantly improves wear resistance and frictional stability [303]. In addition, introducing ionic liquids as structural regulation media during DLC deposition enables the in situ tuning of microstructural evolution during film growth, including sp^2/sp^3 bonding ratio adjustment, nanostructure densification, and residual stress relaxation, thereby fundamentally improving the mechanical stability and tribological response of the coating [300]. This “deposition-process regulation-interfacial lubrication synergy” strategy enables DLC coatings not only to exhibit excellent tribological performance during service, but also to possess an inherently optimized interfacial lubrication foundation established during material synthesis. In practical engineering applications, ionic liquids are typically incorporated as functional additives into base oil systems, forming multi-component solid-liquid lubrication systems. Within such systems, DLC coatings, base oils, and ionic liquids collectively regulate lubrication behavior through multiple mechanisms, including competitive adsorption, molecular rearrangement, and interfacial reactions, thereby establishing a dynamically balanced interfacial lubrication environment. For example,

introducing ionic liquids into water-ethylene glycol or PAO-based oils can significantly enhance the frictional stability and wear resistance of W- or Ag-doped DLC coatings across a wide temperature range and under varying load conditions [302]. Moreover, ionic liquids containing hydroxyl, carboxyl, or other polar functional groups can strengthen hydrogen bonding or weak chemical interactions with the DLC surface, thereby improving interfacial adsorption stability and promoting the formation of continuous and stable tribofilms, further enhancing the overall synergistic lubrication performance [299].

From a multi-scale perspective, the enhancement of lubrication performance in such solid-liquid synergistic systems arises not only from molecular-level adsorption behavior but also involves nanoscale reconstruction of tribofilms and dynamic adjustment of macroscopic contact conditions. During shear, the lubricating film continuously undergoes cycles of formation, rupture, and regeneration, enabling the system to maintain a relatively stable low-friction state under complex operating conditions, such as vacuum, temperature fluctuations, and alternating loads. Overall, the synergistic interaction between DLC coatings and ionic liquids provides an important route for constructing adaptive interfacial lubrication systems and offers new material and mechanistic foundations for reliable lubrication design in extreme aerospace environments.

4 Critical applications and associated challenges of DLC coatings for aerospace applications

In the aerospace domain, components frequently function for protracted periods under extreme conditions, including elevated temperatures, cryogenic environments, high pressures, vacuum, and radiation [6, 26], as well as under high-speed and high-load working conditions [308], which differ markedly from conventional operating scenarios. Such conditions are conducive to accelerated deterioration and degradation, which can potentially compromise the reliability and safety of spacecraft systems [3, 4, 26]. Consequently, enhancing the wear resistance, lubrication efficiency, and environmental adaptability of critical components has emerged as a pivotal challenge in aerospace vehicle design [6]. In this context, DLC coatings have gained prominence as a highly effective solution

due to their exceptional hardness, low friction coefficient, and superior abrasion resistance [26-30].

These coatings offer continuous and reliable surface protection for components operating under extreme conditions, significantly extending their service life and enhancing overall system reliability.

The reduction in maintenance requirements and the mitigation of failure risks contribute to the enhanced operational efficiency and robustness of aerospace systems [6, 26, 27, 29]. However, given the considerable diversity in the functions and service environments of aerospace components, the performance requirements for DLC coatings vary substantially across applications. This section will delve into the application and performance of DLC coatings in the aerospace sector, illustrating their capabilities through representative research examples addressing extreme operational challenges (see Table 1).

Table 1 Tribological performance of DLC coatings on different component surfaces in the aerospace sector.

Aerospace components	Service conditions	Types of DLC coatings	Tribological performance
Gear and spline couplings [309]	High impact, high load, no lubricant	DLC/TiAlN composite coating	The friction coefficient is about 0.5, and the wear rate is about 4×10^{-6} g/N·m.
Valve [310]	Low temperature, frequent actuation, no lubricant	Cr/Cr-W/W-DLC/DLC multilayer coating	Steady state friction coefficients of 0.30 and 0.77 at -50 °C and -100 °C, respectively, while wear rates increase significantly to 4.24×10^{-6} mm ³ /N·m and 9.06×10^{-6} mm ³ /N·m.
Bearing [29]	High temperature, low temperature, vacuum, high load, high gas environment	DLC coatings with different H contents	The average coefficients of friction for the applied load ranges in humid air, dry nitrogen and vacuum are in the range of 0.22 to 0.27, 0.02 to 0.03 and 0.007 to 0.013, respectively.

Others	Ultra-high vacuum				
(Applied to the	and large	DLC/IL	solid-liquid		All coefficients of friction not
moon	alternating	composite	lubrication		greater than 0.12, wear rate at -
exploration	temperature	coating			150 °C maximum, other
project) [7]	differences (-				temperatures are lower
	100 °C to 150 °C)				

4.1 Gear and spline couplings

Gear and spline couplings, as critical heavy-duty transmission components in the aerospace sector, are required to deliver exceptional reliability and prolonged service life under complex operating conditions [309]. However, the commonly employed 18CrNi4A steel is prone to significant wear and failure under high-impact and heavy-load scenarios, particularly in heavy-duty dry-slip conditions, which severely compromise the performance and longevity of the transmission system [311, 312]. To overcome the environmental challenge of severe wear under heavy-load dry-sliding conditions, researchers applied DLC/TiAlN composite coatings to the contact sub-surfaces of transmission components. In this system, the TiAlN interlayer and multilayer composite architecture served as the key performance regulation strategy, improving interfacial bonding strength and load-bearing capability while enhancing wear resistance under high-impact conditions [309]. As illustrated in Fig.13(a), experimental investigations revealed that the DLC/TiAlN composite coating exhibits outstanding wear resistance under high-friction conditions and across various lubricant environments when tested under a 150N load. Furthermore, its superior performance under high-impact and heavy-load conditions was validated through custom-designed spline coupling tests, which unequivocally demonstrated the coating’s capability to provide robust surface protection for heavy-duty transmission components. This development offers an effective solution for enhancing the performance and durability of heavy-duty transmission systems in the aerospace field [309].

4.2 Valve

In the field of aerospace engineering, valves are of paramount importance as critical core components in the delivery, regulation, and shutoff of fuels such as liquid hydrogen and liquid oxygen

[313, 314]. These components often operate under extreme conditions, including deep cryogenic temperatures, high pressures, specialized gas atmospheres, and frequent actuation cycles, where the use of conventional lubricants is severely restricted or entirely infeasible. Such harsh environments render valves highly susceptible to abnormal wear, particularly at the contact interfaces between the spool and the valve seat. This wear can lead to malfunctions such as valve stall, seal failure or unresponsive operation, jeopardizing overall system performance and, in severe cases, resulting in catastrophic mission failure for spacecraft [310, 315]. The challenges posed by high friction from frequent valve actuation and material embrittlement under deep cryogenic conditions render traditional materials and lubrication strategies insufficient to address these issues effectively [310]. Thus, developing innovative solutions to significantly enhance the wear resistance of critical valve components under lubricant-free conditions has become an urgent and pivotal challenge in aerospace engineering.

In recent years, DLC coatings have found increasing applications on valve mating surfaces, owing to their exceptional friction-reducing and wear-resistant properties, showcasing remarkable advantages [310, 316]. In one study shown in Fig.13(b), researchers enhanced the tribological performance of propellant control valve mating surfaces under extreme operating conditions by introducing multilayer gradient DLC coatings with Cr and WC as transition layers. The multilayer gradient architecture, together with the Cr/WC transition layers, effectively relieved residual stress, strengthened interfacial adhesion, and improved the toughness and environmental adaptability of the coating under cryogenic conditions. Tribological experiments conducted over a temperature range from room temperature to cryogenic conditions revealed that the DLC coatings exhibited outstanding friction-reducing and wear-resistant capabilities across a broad temperature spectrum [310]. Moreover, another study demonstrated that DLC coatings significantly reduced friction losses at the cylinder/valve plate interface over a wide range of rotational speeds, thereby effectively improving the overall operational performance of the system [316]. Furthermore, to address the extreme low-temperature and oxygen-

rich service conditions encountered in aerospace liquid-oxygen fuel valves, researchers incorporated W elements into conventional amorphous carbon coatings. During friction, the in-situ formation of tungsten oxides provided enhanced lubricating effects, which effectively improved the tribological behavior and environmental adaptability of the coating under cryogenic oxygen-rich conditions [295].

These findings highlight the potential of DLC coatings as a robust strategy for optimizing the performance of aerospace valves under extreme conditions, while also providing valuable scientific insights and technological support for the advancement of valve design and surface engineering.

4.3 Bearing

Bearings, as critical mechanical components in the aerospace sector, play an indispensable role in supporting and transmitting axial loads. They are extensively employed in high-precision, high-load systems such as spacecraft engines, aviation engines, and satellite control mechanisms [317, 318]. Within the demanding operational environments of aerospace systems, bearings are frequently exposed to extreme conditions, including high temperatures, low temperatures, and vacuum environments. These challenges are particularly pronounced in deep-space exploration missions, where temperature variations can reach hundreds of degrees Celsius, placing exceptionally stringent demands on bearing performance [29]. Bearings must exhibit exceptional stability under severe thermal fluctuations, endure substantial axial loads, and operate reliably under high-speed rotational conditions [29, 169]. Achieving low friction, minimal wear, and extended lifespan in such adverse conditions without reliance on conventional lubricants has emerged as a significant technological challenge in the aerospace field.

To address this challenge, researchers have investigated the application of DLC coatings on bearing contact surfaces. For instance, as displayed in Figure 13(c), one study focused on the insufficient wear resistance and lubrication performance of thrust bearings operating under humid air, dry nitrogen, and vacuum environments [29]. To overcome the strong environmental sensitivity of conventional hydrogenated DLC coatings, researchers employed a high-hydrogen DLC coating incorporating a

silicon transition layer as the key performance regulation strategy. The silicon transition layer effectively enhanced interfacial adhesion and relieved residual stress, while the high-hydrogen DLC structure improved environmental adaptability and maintained stable low-friction behavior under different atmospheric conditions. As a result, the optimized coating exhibited excellent tribological performance and maintained stable operation across the typical load range of thrust bearings in service [29]. Another study focused on spherical plain bearings in a space oscillating mechanism, where hydrogenated DLC coatings were applied to the contact sub-surfaces to enhance load-carrying capacity, impact resistance, wear resistance, and maintenance-free operation. Tribological testing in a vacuum environment revealed that friction-induced heat raised the contact surface temperature to 48 °C, yet this temperature had minimal impact on the tribological performance of the DLC coating, which continued to provide effective friction reduction and wear resistance [319].

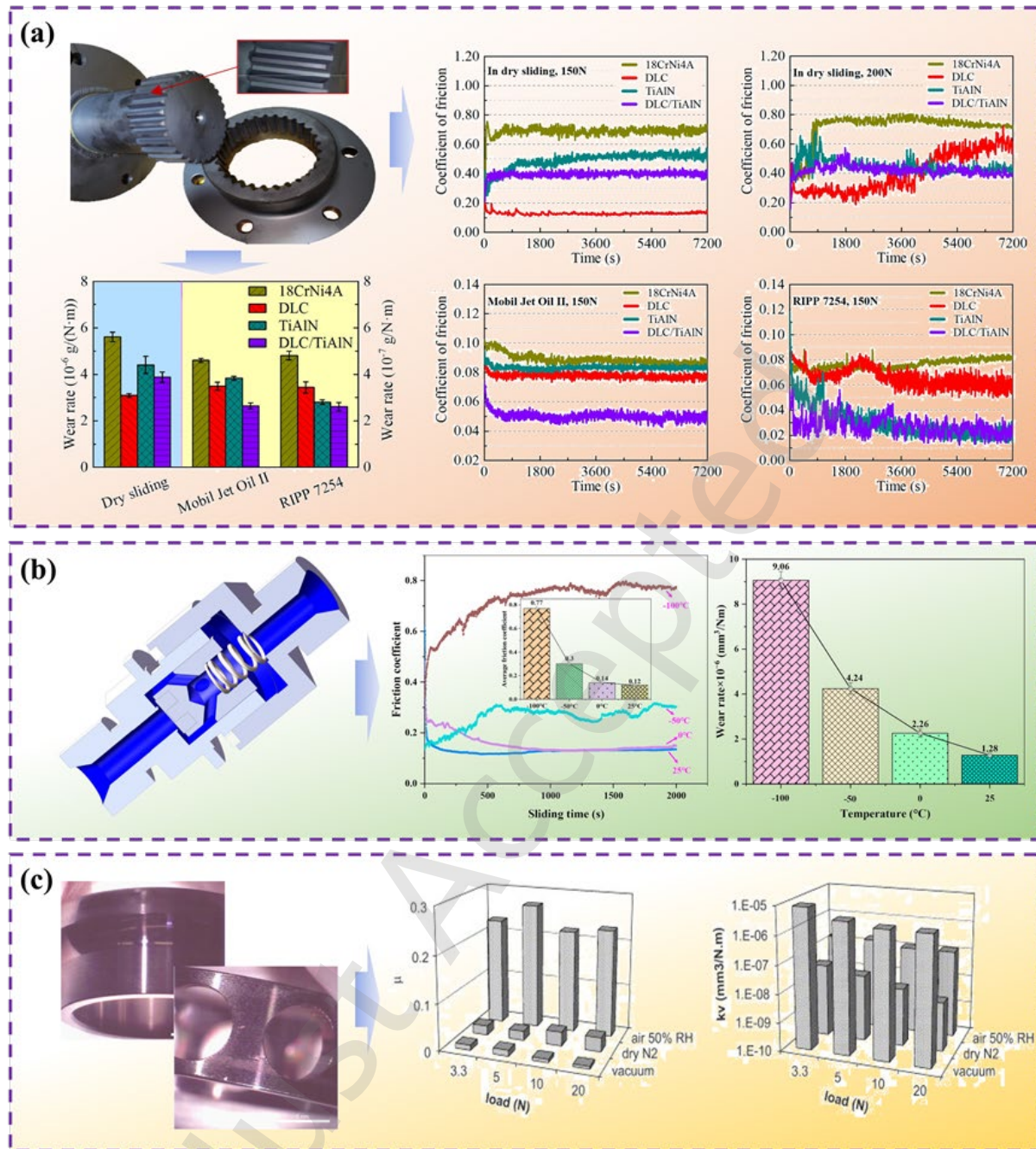


Fig. 13 (a) Tribological properties of DLC/TiAlN composite coatings on spline coupling mating surfaces in aerospace applications [309]. (b) tribological properties of DLC multilayer coatings on valve component mating surfaces in aerospace applications [310]. (c) Tribological properties of hydrogen-containing DLC multilayer coatings on mating surfaces of bearing parts in aerospace applications [29].

These studies highlight innovative solutions to address the tribological challenges faced by bearings under extreme operational conditions. These studies are of particular significance in advancing the

application and development of DLC coating technology in the field of aerospace, representing a substantial step towards the realization of efficient, low-friction, and durable bearing designs, supported by cutting-edge scientific and technological achievements.

4.4 Other applications

DLC coatings have also been widely employed to address tribological challenges in aerospace systems operating under ultra-high vacuum and large temperature fluctuations. For instance, in lunar exploration programs, spacecraft moving components are frequently subjected to extreme environmental conditions, including ultra-high vacuum and alternating temperatures ranging from -100 °C to 150 °C, which can easily induce lubrication failure and severe wear [7]. To overcome these environmental challenges, researchers developed a DLC/ionic liquid (IL) solid-liquid composite lubrication coating, in which the synergistic lubrication strategy combining DLC solid lubrication with ionic-liquid boundary lubrication served as the key performance regulation approach. Friction and wear experiments conducted under high-vacuum and alternating-temperature conditions revealed that the coating exhibited the lowest and highest friction coefficients at 100 °C and -100 °C, respectively. Further analysis demonstrated that slight graphitization of the DLC surface at elevated temperatures, together with the excellent fluidity, reactivity, spreadability, and self-repairing capability of the ionic liquid, jointly contributed to enhanced tribological performance and environmental adaptability under thermal-vacuum conditions [7]. Another study similarly demonstrated that the superior tribological behavior of DLC/IL composite lubrication coatings under room-temperature vacuum conditions originated not only from the low viscosity and high fluidity of the ionic liquid, but also from the synergistic lubrication mechanism between the DLC coating and the liquid lubricant boundary film [320].

Furthermore, to address friction and wear problems in aerospace drive mechanisms operating under high-vacuum conditions, researchers developed a grease/DLC solid-liquid synergistic lubrication system [321]. In this system, the synergistic coupling between the DLC coating and the lubricating

grease acted as the key performance regulation strategy for improving lubrication stability and environmental adaptability under vacuum conditions. Experimental results showed that the hybrid grease not only provided effective lubrication, but also significantly enhanced the tribological performance of the DLC coating, particularly when an optimized grease-to-DLC ratio was employed. Under low-load conditions, the excellent tribological behavior mainly originated from the synergistic interaction between the DLC coating and the fluid lubrication film, while under more severe operating conditions, the cooperative lubrication effect between grease and the DLC coating became even more pronounced. These studies further demonstrate that designing targeted synergistic lubrication strategies according to specific aerospace environmental challenges is essential for improving the long-term reliability of DLC coatings under extreme service conditions.

5 Conclusions and future outlook

5.1 Conclusions

In this article, a systematic exposition is first presented on the pronounced environmental sensitivity of the tribological performance exhibited by DLC coatings under diverse service conditions. This is followed by a comprehensive review and critical analysis of the principal strategies and underlying mechanisms currently employed to modulate the tribological behavior of DLC coatings. Finally, by integrating representative application cases in aerospace and other high-end engineering domains, an insightful retrospection and forward-looking perspective is offered on the practical engineering applications and associated challenges of DLC coatings. The primary conclusions that can be drawn from this analysis are as follows:

(1) The tribological performance of DLC coatings in service is highly sensitive to external environmental factors such as temperature and gaseous atmosphere. The interfacial interactions between the coating and the surrounding medium frequently give rise to complex and competing friction-induced chemical reactions, which in turn play a pivotal role in dictating the tribological response. Therefore, to ensure the efficient and reliable operation of DLC coatings under specific

working conditions, it is imperative that their design strategies holistically consider the chemical composition, reactivity, and thermal evolution of the target environment. On this basis, the construction of interface structures and compositional gradient systems endowed with temperature responsiveness and atmospheric adaptability becomes essential to achieving environmentally adaptive control over interfacial tribological behavior.

(2) Multilayer structure design, heterogeneous element doping, and surface morphology engineering are the three principal strategies for regulating the tribological performance of DLC coatings under extreme environments. Multilayer and gradient architectures can effectively relieve residual stress, enhance interfacial adhesion, and improve resistance to crack propagation, thereby enhancing coating stability and load-bearing capacity. Heterogeneous element doping can regulate the sp^2/sp^3 bonding structure, tribochemical activity, and thermal stability of DLC coatings, enabling improved environmental adaptability and friction-reduction behavior under different gaseous atmospheres and temperature conditions. In addition, surface morphology engineering can optimize interfacial contact states, debris transfer behavior, and lubricating film formation, thereby reducing friction and wear under complex operating conditions. These regulation strategies exhibit distinct modification mechanisms but collectively provide effective pathways for improving the reliability and long-term service performance of DLC coatings in aerospace applications.

(3) The most significant barrier to the widespread adoption of DLC coatings in aerospace applications is the presence of extreme service conditions, particularly in harsh environments such as high and low temperatures, vacuum, and high-speed conditions, where the coating's performance can be substantially compromised. Furthermore, the service functions and operating conditions of different components vary considerably, rendering the customization of coatings for each specific application a complex and challenging endeavor.

5.2 Future outlook

Despite the exploration of DLC coatings in the aerospace industry, their utilization remains limited. The primary reason for this is that the performance of these coatings in extreme environments has yet to fully meet the stringent requirements for high-performance materials in this field. In particular, the durability and reliability of DLC coatings are challenged under harsh operating conditions, such as elevated and low temperatures, radiation, and complex atmospheres. However, with the continuous enhancement of material performance standards, the potential of DLC coatings in this domain is gradually being realized. In the future, it is anticipated that DLC coatings will find broader applications in the following areas:

(1) Optimization of micro- and nanostructures

The integration of micro- and nanofabrication technologies has enabled the development of DLC coatings, which facilitate precise surface control on the complex components of aerospace vehicles. This, in turn, enhances their surface properties, including smoothness, hardness, and surface energy. The adaptability of these coatings to diverse environments is further enhanced through the optimization of factors such as thickness, porosity, and the doping of heterogeneous elements.

(2) Self-healing DLC-based interfacial functional materials

During prolonged operation, spacecraft coatings inevitably suffer from fatigue accumulation, microcrack propagation, and abrasive particle damage. Conventional DLC coatings, however, lack intrinsic self-healing capabilities to recover from such micro-damages, thereby limiting their long-term protective performance. Future developments may focus on integrating advanced material systems such as microcapsule embedding technology, metal-organic frameworks (MOFs), and multilayered interfacial nanostructures to engineer self-healing DLC coatings with comprehensive functionalities including damage recognition, responsive release, and structural reconstruction. This approach enables active sensing and intelligent repair of in-service micro-damages, thereby substantially enhancing the coating's service life and reliability.

(3) Multifunctional integrated DLC composite coating systems

To meet the urgent demands for lightweight and multifunctional integration in aerospace components, DLC coatings are evolving from traditional single-function wear protection layers into multifunctional composite interfacial systems that integrate friction reduction and wear resistance, thermal management, electromagnetic shielding, and corrosion resistance. By engineering layered composite architectures, such as DLC/metal interlayer/DLC sandwich structures, or incorporating functional fillers like thermally conductive ceramic particles and two-dimensional conductive nanosheets, synergistic regulation of thermal, electrical, and mechanical fields can be achieved. This approach endows the coating systems with enhanced interfacial coupling performance and multi-field adaptability, thereby ensuring efficient and stable service under complex aerospace operating conditions.

(4) Environmentally adaptive intelligent coating systems

Faced with the severe temperature fluctuations, complex atmospheric variations, and frequent mechanical contacts encountered during spacecraft in-orbit operations, the conventional single-function physical protection offered by DLC coatings is increasingly inadequate to meet the demands of multi-physics coupled service conditions. Future advancements are anticipated through the integration of multi-scale structural design and intelligent responsive materials to develop smart DLC coating systems endowed with self-sensing, self-regulating, and environmentally adaptive capabilities, enabling dynamic control and optimization of tribological behavior under complex service environments.

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