

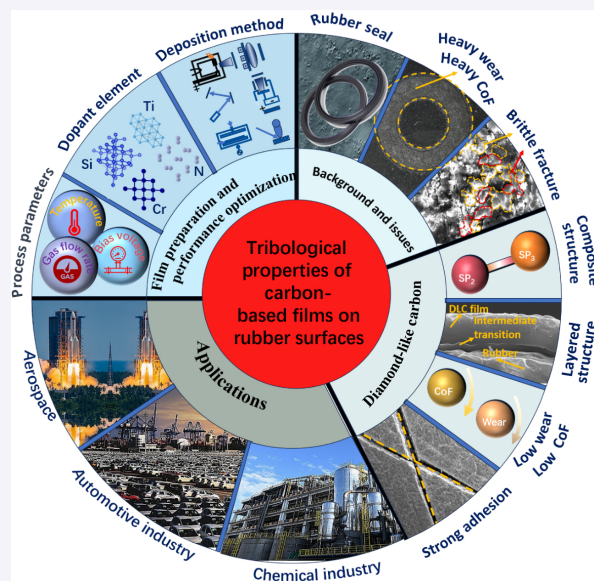
A review on the tribology of carbon coatings on rubber

Weiye Li^{1,2}, Li Chen², Bin Zhang¹✉

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ABSTRACT: Rubber is widely used as a crucial sealing material in the aerospace, petrochemical, and automotive industries to prevent contaminants from entering enclosures and lubricants from leaking. However, severe wear and friction occur during sliding motion, which are major causes of seal failure and significantly impact the safety and service life of equipment. To increase the wear resistance of rubber surfaces, diamond-like carbon (DLC) coatings have been extensively studied because of their low friction coefficient, high hardness, excellent wear resistance, and chemical inertness. The hardness, elasticity, and adhesion of the coating can be effectively controlled by adjusting the deposition parameters. This allows the film to accommodate the deformation of the soft rubber substrate, preventing delamination while avoiding thermal degradation or damage to the rubber. Additionally, the chemical composition of carbon-based films, which primarily consist of carbon and hydrogen, is compatible with rubber, ensuring strong interfacial adhesion. In this paper, research progress on the tribological properties of carbon-based films for rubber surface modification over the past two decades is reviewed. In contrast to previous reviews that focused primarily on the general applications and fundamental properties of DLC coatings, this work delves into the distinctive advantages of DLC coatings on rubber surfaces under various deposition conditions. This work explores the underlying mechanisms for friction reduction and wear resistance enhancement while also identifying critical research gaps and proposing future directions for the field.

KEYWORDS: rubber; carbide films; diamond-like carbon (DLC) films; tribological properties; mechanical properties; deposition conditions



1 Introduction

With the development of modern industrial civilization, many sealing components are present in almost all mechanical equipment. The primary function of these components is to prevent fluid leakage and the ingress of contaminants into the machinery [1]. Once a sealing component fails and leakage occurs, it can lead to material loss, equipment damage, or even hazardous situations such as explosions. Owing to their excellent elasticity and wear resistance, rubber materials are widely used in sealing applications [2–6]. However, in practical use, friction and wear on rubber surfaces significantly affect their service life and system efficiency [7, 8]. To address the short service life of rubber sealing components, the development of surface modification technologies capable of effectively reducing the friction coefficient and enhancing wear resistance is particularly important. As an

effective modification approach, rubber surface modification involves the deposition of a thin film with specialized properties onto the rubber surface, significantly improving its tribological performance and enabling its application under complex operating conditions.

In recent years, significant progress has been made in thin-film technologies for rubber surface modification. These films can be fabricated from various materials, including polymers, metals, ceramics, and carbon-based materials, each offering distinct properties and application advantages. For example, polymer films exhibit excellent flexibility and corrosion resistance [9–11], and metal films provide high strength and electrical conductivity [12, 13]. Carbon-based films have attracted widespread attention because of their low friction coefficient, high hardness, and excellent chemical stability [14–18]. However, the application of thin-film technology on rubber surfaces also faces several challenges, such

¹ Key Laboratory of Science and Technology on Wear and Protection of Materials, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, China. ² College of Petroleum and Chemical Engineering, Lanzhou University of Technology, Lanzhou 730050, China.

✉ Corresponding author. E-mail: bzhang@licp.cas.cn

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as the adhesion strength between the film and the rubber substrate, the uniformity and stability of the film, and its ability to maintain performance under various operating conditions. Additionally, the fabrication process and associated costs are critical factors influencing its widespread application. Therefore, an in-depth investigation into the tribological properties of thin films on rubber surfaces and their modification mechanisms is highly important for enhancing the service life and performance of rubber materials.

This paper reviews the research progress on the tribological properties of thin films on rubber surfaces, with a particular focus on carbon-based films. First, the current applications of various thin-film materials and rubber surface modification coatings are introduced. Then, the tribological properties of the modified rubber surface films are analyzed, followed by a discussion of their mechanical properties. Finally, future research directions and potential application areas are proposed. Through systematic analysis and discussion, this review aims to provide theoretical support and practical guidance for the research and application of thin films on rubber surfaces.

2 Classification and preparation of rubber surface films

Figure 1 provides an overview of conventional rubber substrates and common solid lubricant materials. Conventional rubber materials include natural rubber (NR) [19], styrene-butadiene rubber (SBR) [20], nitrile butadiene rubber (NBR), butadiene rubber (BR) [21], ethylene propylene diene monomer rubber (EPDM) [22], fluoroelastomers (FKMs) [23], and polyurethane rubber (PU) [24]. Common soft metals used as solid lubricant matrices include Ag [25, 26], Pb [27, 28], Au [29, 30], Sn [31], and Cu [32]. The polymers used for this purpose include polytetrafluoroethylene (PTFE) [33–35] and polyimide [36, 37]. Layered solid materials include MoS_2 [38], WS_2 [39], graphite [40], hexagonal boron nitride (HBN) [41], and GaSe [42]. Carbides include TiC [43], WC [44], and CrC. Nitrides include TiN [8, 45], CrN [46], and BN [47]. The structural characteristics, advantages, and limitations of the aforementioned materials are summarized in Table 1. Table 2 compares the chemical properties, elastic moduli, and friction coefficients of the different solid

lubricant matrices. In practical applications, these solid lubricant matrices have certain limitations. For example, soft metal materials do not exhibit low-temperature brittleness [48] but are prone to plastic deformation and have poor load-bearing capacity. PTFE polymer materials have a low friction coefficient and excellent wear resistance [49–51]; however, they are difficult to process and shape, and their manufacturing cost is relatively high [51]. Carbide and nitride materials possess high hardness, high melting points, and good chemical stability, providing excellent wear resistance [52–60], but their films tend to be brittle. However, it is also necessary to consider whether the aforementioned solid lubricant materials can be well matched with rubber. Therefore, alternative coatings with superior properties need to be explored. Owing to their low friction, high hardness, and excellent wear resistance [16, 61–64], diamond-like carbon (DLC) films have been proven to significantly enhance the tribological performance of rubber surfaces.

2.1 Overview of carbon films

Carbon is ubiquitous in nature and serves as the fundamental

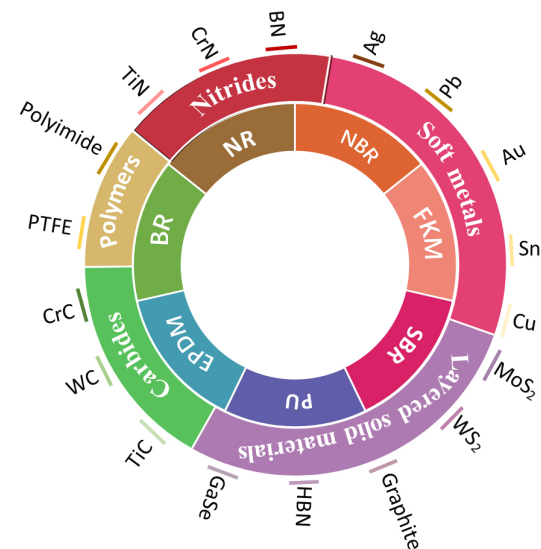


Fig. 1 Conventional rubber substrates and common solid lubricant materials.

Table 1 Solid lubricating materials: structure, advantages, and limitations

Solid lubricant material	Structural feature	Advantage	Limitation
Soft metals (e.g., Ag, Pb, Au, In, Sn, Cu)	Face-centered cubic structure, prone to intergranular slip [65–68]	No low-temperature brittleness, strong bonding with metal substrates; good plasticity, low friction coefficient, and excellent wear resistance [69, 70]	Poor load-bearing capacity, prone to plastic deformation [71]; susceptible to oxidation at high temperatures [72]
Polymers (e.g., PTFE, polyimide)	Fully symmetric or highly symmetric molecular structure, low polarity or nonconjugated structure [73, 74]	Extremely low friction coefficient, excellent wear resistance, superior sealing and lubrication properties, good heat [75, 76], and chemical solvent resistance [77, 78]	Low mechanical strength, limited load-bearing capacity [79]
Layered solid materials (e.g., MoS_2 , WS_2 , graphite, h-BN, GaSe)	Hexagonal close-packed structure, weak van der Waals forces between layers [80–83]	Low friction coefficient and good wear resistance [84, 85]	Prone to moisture absorption, poor thermal stability, susceptible to corrosion [86]
Carbides (e.g., TiC, WC, CrC)	Crystalline solids, high hardness, and high melting point [87–90]	High hardness, chemically stable [91], excellent wear resistance [92], and suitable for high-temperature [93]	Brittle, poor impact resistance [94, 95]; difficult to process, high costs
Nitrides (e.g., TiN, CrN, BN)	Crystalline solids, high hardness and high melting point [57, 96–98]	High-temperature resistance, excellent wear resistance [99], and good thermal shock resistance	Brittle, poor impact resistance [100], difficult to process, and high costs
Diamond-like carbon (e.g., a-C, a-C:H, Ta-C, Ta-C:H, CN _x)	Network structure formed by sp and sp ² hybridized carbon atoms covalently bonded with hydrogen or nitrogen [101]	Low friction coefficient, excellent wear resistance [102], tunable mechanical properties, and suitable for various working conditions	High internal stress, prone to delamination [103], complex preparation process, and high costs

Table 2 Chemical, mechanical, and tribological properties of different solid lubricant matrices

Thin-film material	Chemical property	Chemical resistance	Young's modulus (GPa)	Coefficient of friction
Ag	Face-centered cubic structure and easily oxidized [104]	Good alkali resistance and easily corroded by acid [105]		0.5–1.0 [106]
Cu	Face-centered cubic structure and moderate corrosion resistance [107]	Good corrosion resistance [108]	110–128 [109]	0.6–1.2 [106]
PTFE	Polymer with a linear chain structure and high chemical inertness [110]	Resistant to strong acids, strong alkalis and organic solvents [111]	0.5–1.5 [112]	0.05–0.3 [49, 113, 114]
MoS ₂	Layered structure, weak van der Waals forces [115], and good lubricity [82]	MoS ₂ /rGO/NiS-5 is resistant to acidic environments but unstable under neutral and alkaline conditions [116]		0.05–0.15 [117, 118]
TiC	Cubic structure [119] and high hardness [120, 121]	Resistant to acids, bases, and oxidation [121]	400–450 [122]	0.3–0.5 [123, 124]
CrC	Hexagonal/cubic structure and high wear resistance [125]	Resistant to acids and high-temperature oxidation [126]		0.4–0.6 [127]
TiN	Cubic structure [128, 129] and high hardness [130]	Resistant to acids, bases, and corrosion [131]	250–600 [132]	0.4–0.6
CrN	Cubic structure and good corrosion resistance [133, 134]	Superior to TiN; resistant to high-temperature oxidation [135]	300–400 [136]	0.5–0.7
DLC	Amorphous/graphite-like structure and high sp ³ content [137, 138]	Resistant to acids, bases, and organic solvents [139]	200–1,000 [140]	0.05–0.2 [141]

framework for all living organisms. It exists on Earth in various forms, including stable graphite crystals, relatively unstable diamond crystals, and a wide range of hydrocarbons. Typically, amorphous carbon (a-C) films with a structure resembling that of diamond are referred to as DLC films, whereas those with a graphite-like structure are known as graphitic-like carbon films. Films with a polymer-like structure are termed polymer-like carbon films. All these types can be categorized as carbon-based thin films. DLC films are a type of amorphous carbon thin film with a diamond-like structure [142–144]. Research on DLC films originated from successful deposition by Aisenberg and Chabot in the 1970s [145]. However, extensive studies on their mechanical and tribological properties did not emerge until the 1990s. At room temperature, carbon atoms primarily bond through three hybridization states: sp, sp², and sp³ [146]. In 1993, Jacob and Möller [147] summarized literature and classified DLC films on the basis of sp³ hybridization, sp² hybridization, and hydrogen content, leading to the first ternary phase diagram of DLC films. According to the ternary phase diagram of amorphous carbon, when carbon atoms bond entirely in the sp² state, pure graphite crystals are formed, whereas pure sp³ bonding results in the formation of pure diamond. In DLC films, both sp² and sp³ bonding configurations coexist, resulting in properties and structures that lie between those of diamond and graphite [148]. As the ratio of sp³ to sp² bonding in the DLC film varies, its characteristics shift accordingly, making it more similar to either diamond or graphite. On the basis of the bonding configurations and proportions of carbon atoms, as well as structural properties such as hardness and hydrogen content, DLC films can be broadly classified into four types [149]: a-C films, hydrogenated amorphous carbon (a-C:H) films [150], tetrahedral amorphous carbon (Ta-C) films, and hydrogenated tetrahedral amorphous carbon (Ta-C:H) films [149]. Compared with a-C films, a-C:H films are relatively more flexible and exhibit lower internal stress. Additionally, a-C:H films generally demonstrate lower friction, as hydrogen can effectively passivate dangling σ bonds. Moreover, some hydrogen atoms in a-C:H films do not initially bond with other atoms, leading to the formation of a porous structure [151]. This porous structure enhances the flexibility of the film, allowing it to better accommodate the deformation of the rubber substrate. Moreover, the C-H bonds in a-C:H films are covalent (stronger than individual C-C bonds), making them difficult to remove

from the surface unless subjected to high-temperature friction tests [152]. It is evident that a-C:H films are a highly suitable choice for deposition on rubber surfaces.

2.1.1 Preparation of carbon films

As shown in Fig. 2(a), this figure intuitively summarizes several commonly used methods for depositing carbon-based films on rubber surfaces. The current deposition techniques for carbon films on rubber surfaces include plasma-enhanced chemical vapor deposition (PECVD) [153], pulsed laser deposition (PLD) [154], vacuum cathodic arc deposition (VCAD) [155], vacuum cathodic arc deposition [156], ion beam deposition [157], and T-shaped filtered arc deposition (T-FAD) [158, 159]. Specific details about these methods are summarized in Table 3. Cross-disciplinary collaboration is crucial for advancing the preparation technology of carbon films on rubber surfaces. Material science lays the theoretical foundation for film design and preparation. Chemistry delves into the mechanisms of interaction between the film and the rubber matrix. Mechanical engineering optimizes film performance on the basis of actual working conditions. Additionally, computer science, particularly AI and big data technologies, provides robust support for optimizing film structures and predicting performance. Through such multidisciplinary integration, breakthroughs in existing technological bottlenecks are expected to be achieved, and high-performance carbon film preparation technologies for rubber surfaces can be developed, effectively addressing the deficiencies of current carbon film preparation methods.

2.1.2 Research progress on DLC coatings on rubber

Figure 2(b) illustrates the research progress of DLC coatings on rubber in various materials and application fields in recent years. The figure presents statistical data on the advancements and key findings in the application of DLC coatings for rubber surface lubrication from 2000 to the present. In 2007, Yoshida et al. [160] investigated the deposition of low-friction, high-hardness DLC films on silicone rubber via femtosecond PLD technology. The results indicated that the deposition rate on silicone rubber was higher than that on silicon wafers, and the films presented a higher sp³ content along with a rough textured structure. These characteristics significantly improved the friction performance of the silicone rubber. The DLC/silicone rubber structure

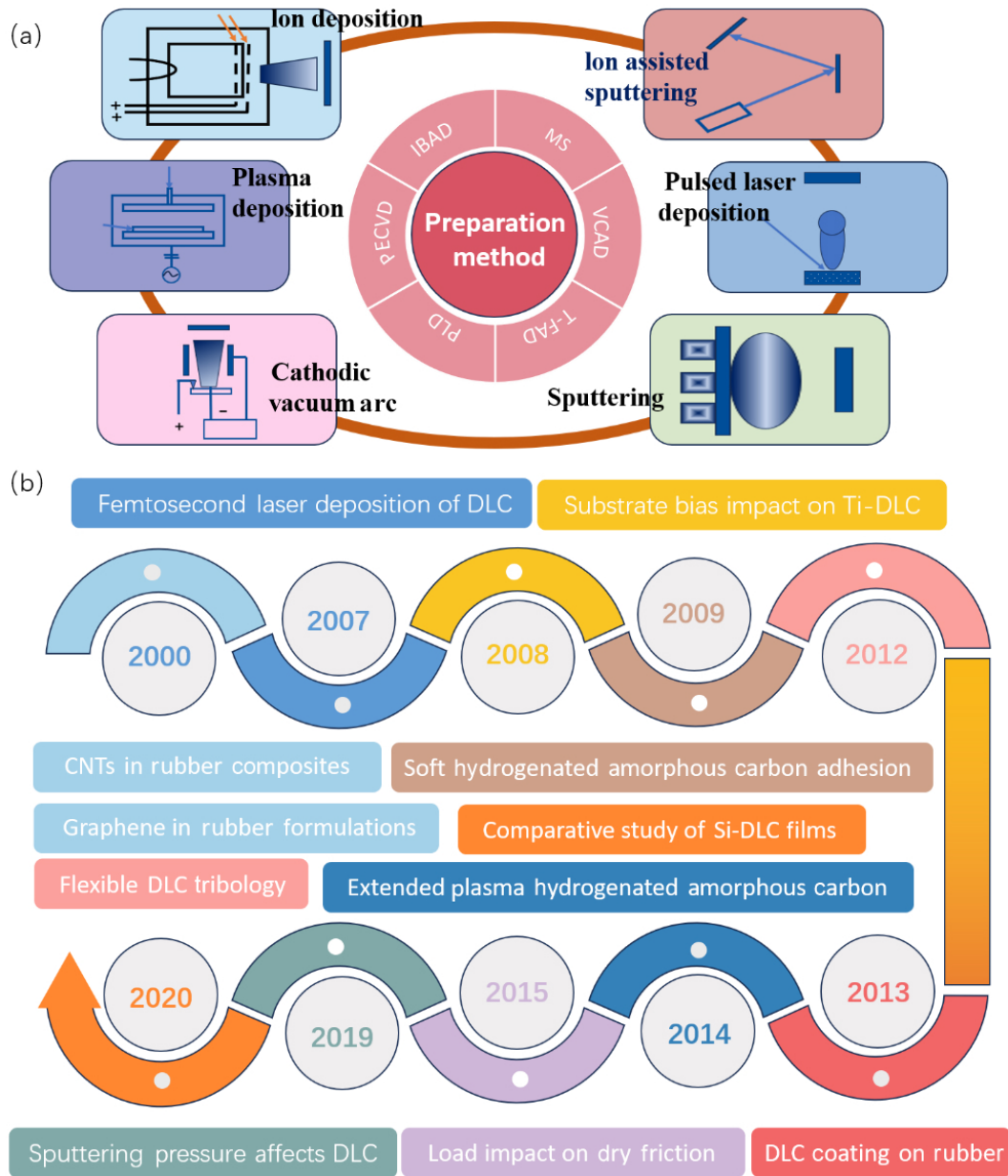


Fig. 2 (a) Deposition methods of DLC films on rubber sealing materials. (b) Research progress on DLC coatings on rubber in various materials and applications.

Table 3 Methods, principles, advantages, and disadvantages of carbon thin film deposition on rubber surfaces

Method	Principle	Advantage	Disadvantage
Ion beam assisted deposition (IBAD)	An ion source bombards the target to generate an ion beam, which is accelerated by an electric field for deposition; an auxiliary ion beam further optimizes film performance	Low contamination, controllable parameters; high sp^3 content and smooth surface	Limited deposition size, low deposition rate, and high equipment cost
Magnetron sputtering (MS)	A magnetic field enhances sputtering, causing target atoms to deposit and form a film	High purity, excellent uniformity; suitable for large-scale production	Low target utilization, potential substrate damage
VCAD	Arc evaporation of the target produces high-energy ions that deposit onto the substrate	High film density, wear resistance, corrosion resistance, and high-temperature resistance	Prone to droplet contamination, complex process parameters
T-FAD	An enhancement of VCAD with a magnetic filtering device to remove large particles and neutral species	Higher film purity and fewer droplet defects	Low deposition rate, more complex equipment
PLD	A short-pulse laser ablates the target, and the resulting plasma plume deposits directionally	Precise stoichiometry and high hardness	Limited deposition area, high energy consumption, and particle residues
PECVD	Deposition using plasma-activated gas at low temperatures	Low-temperature process, dense and pore-free films suitable for complex substrates	High hydrogen content and poor thermal stability

demonstrated in this study has potential for applications across various industrial fields. In 2008, Bui et al. [161] studied the effect of substrate bias on Ti-DLC coatings, specifically investigating its influence on the deposition rate, structure, hardness, and tribological performance of Ti-DLC films on hydrogenated nitrile butadiene rubber (HNBR). The results showed that as the substrate bias increased from -50 to -300 V, the deposition rate decreased from 15.5 to 7.1 nm/min, while the columnar structure of the coating was suppressed, and the segmented morphology became finer. Additionally, the hardness increased from 9.4 to 16.2 GPa, and the friction coefficient significantly decreased. Notably, at a bias of -300 V, the lowest friction coefficient of 0.21 was achieved, indicating that a higher substrate bias can substantially enhance the performance of the coating. In 2012, Pei et al. [162] investigated the tribological behavior of flexible DLC films on HNBRs and the effect of viscoelastic deformation of the rubber substrate on friction. By utilizing the thermal mismatch between the DLC film and the rubber substrate, a dense crack network was formed, imparting flexibility to the DLC film. This study examined the influence of the microsegmented structure size of a DLC film on its frictional performance. Furthermore, friction tests and theoretical analysis were conducted to explore the impact of the viscoelasticity of the rubber substrate on the tribological behavior of the DLC-coated system. In 2013, Pei et al. [163] investigated the coating of flexible DLC films on rubber. By exploiting the thermal mismatch between the DLC film and the rubber substrate, a dense crack network was induced, enhancing the flexibility of the film and reducing the friction coefficient. The experimental results indicated that smaller DLC film segment sizes significantly lowered the friction coefficient of rubber and improved its wear resistance. This study provides a general design

guideline for low-friction coatings on dynamic rubber seals (Fig. 3). In 2019, Liu et al. [164] investigated the effects of sputtering pressure on the surface morphology, structure, wettability, and tribological properties of DLC films prepared by magnetron sputtering on rubber substrates. The study revealed that increasing the sputtering pressure influences the surface roughness, chemical bonding structure, and surface energy of DLC films, thereby affecting their tribological performance. In 2020, Qiang et al. [165] investigated the microstructure, adhesion, and tribological properties of silicon-doped DLC films deposited on nitrile butadiene rubber and compared them with those of undoped DLC films. By first depositing a silicon interlayer on NBR and then applying the Si-DLC film, the influence of the interlayer thickness on the microstructure, adhesion, and frictional behavior of the coating was systematically studied. This research provides significant experimental and theoretical insights into the deposition of high-performance DLC films on rubber substrates.

Through previous research, a key topic in the field of rubber—surface modification—has been revealed. The most important consideration is the adjustment of the microstructure of the film, which involves adjusting the sp^2/sp^3 carbon bond ratio. This adjustment plays a key role in optimizing the tribological and mechanical properties of the film. In this context, emerging AI-aided design technologies show great potential. Machine learning algorithms can predict how different carbon bond ratios affect thin film properties, enabling them to quickly identify the optimal thin film structural parameters. Another key topic is the interfacial bonding mechanism between the film and the rubber matrix. This mechanism is essential for enhancing the adhesion and durability of the film. Molecular dynamics simulations can be used to study the atomic-level interactions between thin films and rubber

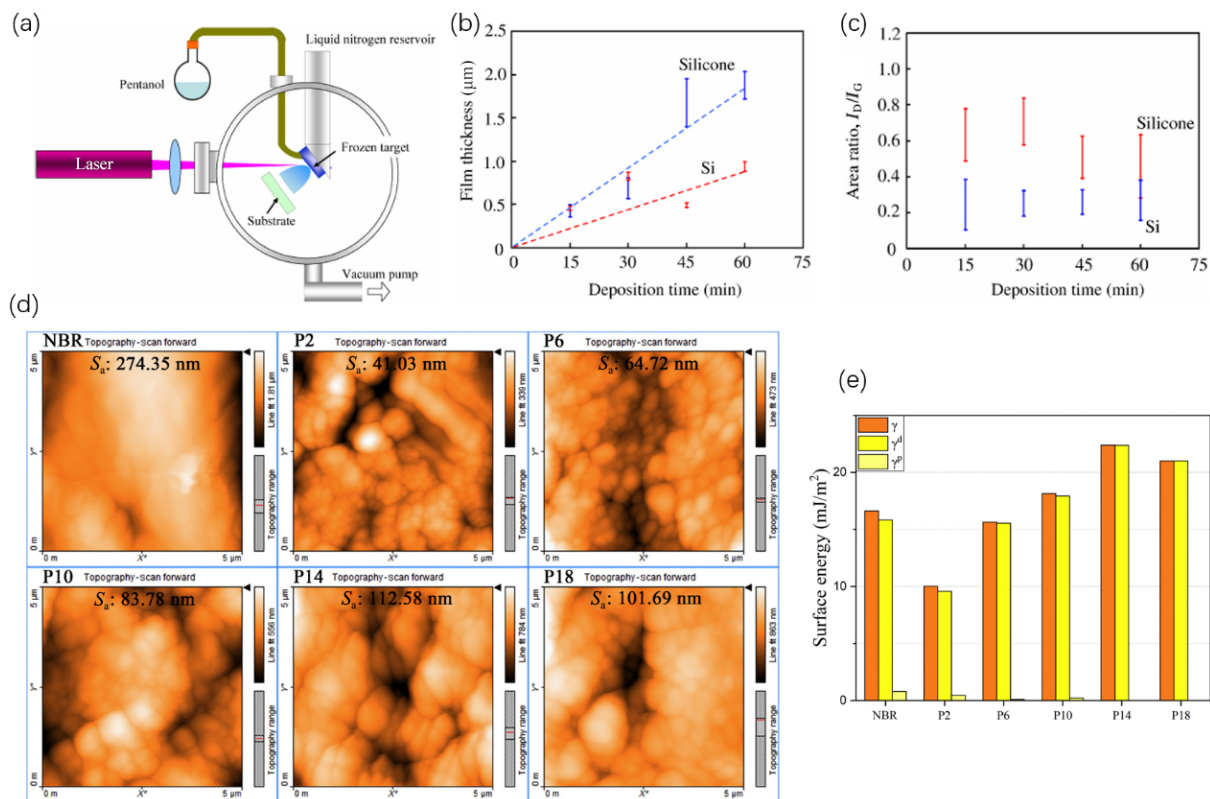


Fig. 3 (a) A side view of the arrangement used in the present PLD experiment. (b) Film thickness as a function of the deposition time on two different substrates. (c) I_D/I_G Raman peak area ratio as a function of the deposition time. (d) AFM micrographics of NBR and DLC films deposited on NBR under different sputtering pressures. (e) Total and component surface energies of NBR and DLC surfaces deposited under different sputtering pressures (SPs), calculated via the OWRK method. Reproduced with permission from Ref. [160] for (a–c), © IOP Publishing Ltd 2007; Ref. [164] for (d) and (e), © Elsevier B.V. 2019.

matrices in depth, thus providing theoretical guidance for experimental design. On the basis of the aforementioned properties, the tribological performance of carbon-based films on rubber surfaces is further explored in Section 3.

3 Tribological properties of rubber surface films

Table 4 summarizes research from a wide range of literature on the coating of different types of thin films on various rubber

substrates, encompassing key information such as ball-on-disk test parameters, friction environments, and friction coefficients. By comparing the data from different studies, it is evident that DLC films on rubber surfaces exhibit outstanding performance in reducing the friction coefficient, significantly enhancing the tribological properties of rubber (Fig. 4). This finding further confirms the superiority of DLC films in improving the wear resistance of rubber and reducing frictional losses, providing

Table 4 Friction conditions and friction coefficients of different films on different rubbers

Sample		Tribological property						Ref.
Substrate	Film	Counter part	Diameter (mm)	Load (N)	Speed (cm/s)	Lubr	CoFs	
NBR	DLC	WC-Co	5	1	10	Dry	0.34	[166]
NBR	Si-DLC	WC-Co	5	1	10	Dry	0.34	[166]
NBR	DLC + Si-C	WC-Co	5	1	10	Dry	0.36	[166]
NBR	Si-DLC + Si-C	WC-Co	5	1	10	Dry	0.38	[166]
NBR	DLC	WC-Co	5	1	10	Wet	0.52	[166]
NBR	Si-DLC	WC-Co	5	1	10	Wet	0.45	[166]
NBR	DLC + Si-C	WC-Co	5	1	10	Wet	0.44	[166]
NBR	Si-DLC + Si-C	WC-Co	5	1	10	Wet	0.42	[166]
NBR	DLC	WC-Co	5	5	10	Dry	0.35	[166]
NBR	Si-DLC	WC-Co	5	5	10	Dry	0.4	[166]
NBR	DLC + Si-C	WC-Co	5	5	10	Dry	0.46	[166]
NBR	Si-DLC + Si-C	WC-Co	5	5	10	Dry	0.57	[166]
NBR	DLC	WC-Co	5	5	10	Dry	0.25–0.45	[167]
NBR	Si-DLC	WC-Co	5	5	10	Dry	0.25–0.6	[167]
NBR	DLC + Si-C	WC-Co	5	5	10	Dry	0.25–0.6	[167]
NBR	Si-DLC + Si-C	WC-Co	5	5	10	Dry	0.25–0.6	[167]
NBR	Si-DLC	GCr15	6	3	12.5	Dry (humidity 27%±1%)	0.25–0.29	[168]
NBR	Si-DLC	GCr15	6	3	12.5	Dry (humidity 27%±1%)	0.23–0.27	[165]
NBR	DLC	304		1–10		Dry	0.52–1.05	[169]
NBR	GLC	GCr15		5–10		Wet	0.26	[170]
NBR	Ti-C	GCr15		5–10		Wet	0.15–0.17	[170]
NBR	TiC-C	GCr15		5–10		Wet	0.15–0.17	[170]
NBR	TiN-C	GCr15		5–10		Wet	0.26–0.28	[170]
NBR	DLC	ZrO ₂	1.6	0.3	4	Dry	0.5	[171]
NBR	N-DLC/DLC	ZrO ₂	1.6	0.3	4	Dry	0.2–0.4	[171]
NBR	Ti-DLC	ZrO ₂	1.6	0.3	3	Dry	0.18	[172]
NBR	DLC	GCr15	6	3	8.4	Dry (humidity 25%±3%)	0.2	[173]
NBR	DLC	GCr15	4	10	8.4	Dry (humidity 25%±1%)	0.2–0.28	[174]
HNBR	DLC	100Cr6	6	1	10	Dry (humidity 35%±1%)	0.10–0.17	[162]
HNBR	DLC	100Cr6	6	3	10	Dry (humidity 35%±1%)	0.13–0.19	[162]
HNBR	DLC	100Cr6	6	5	10	Dry (humidity 35%±1%)	0.14–0.21	[162]
HNBR	DLC	100Cr6	6	1	10	Dry (humidity 35%±2%)	0.12–0.13	[175]
PU	DLC	100Cr6	10	1	10	Dry	0.6	[176]
PU	F-DLC	100Cr6	10	1	10	Dry	0.3–0.6	[176]
FKM	DLC	GCr15	6	1	3	Dry	0.5	[177]
FKM	DLC	304		1–10		Dry	0.58–1.05	[169]
EPDM	DLC	GCr15	6	1	3	Dry	0.2	[177]
EPDM	DLC	304		1–10		Dry	0.4–1.05	[169]

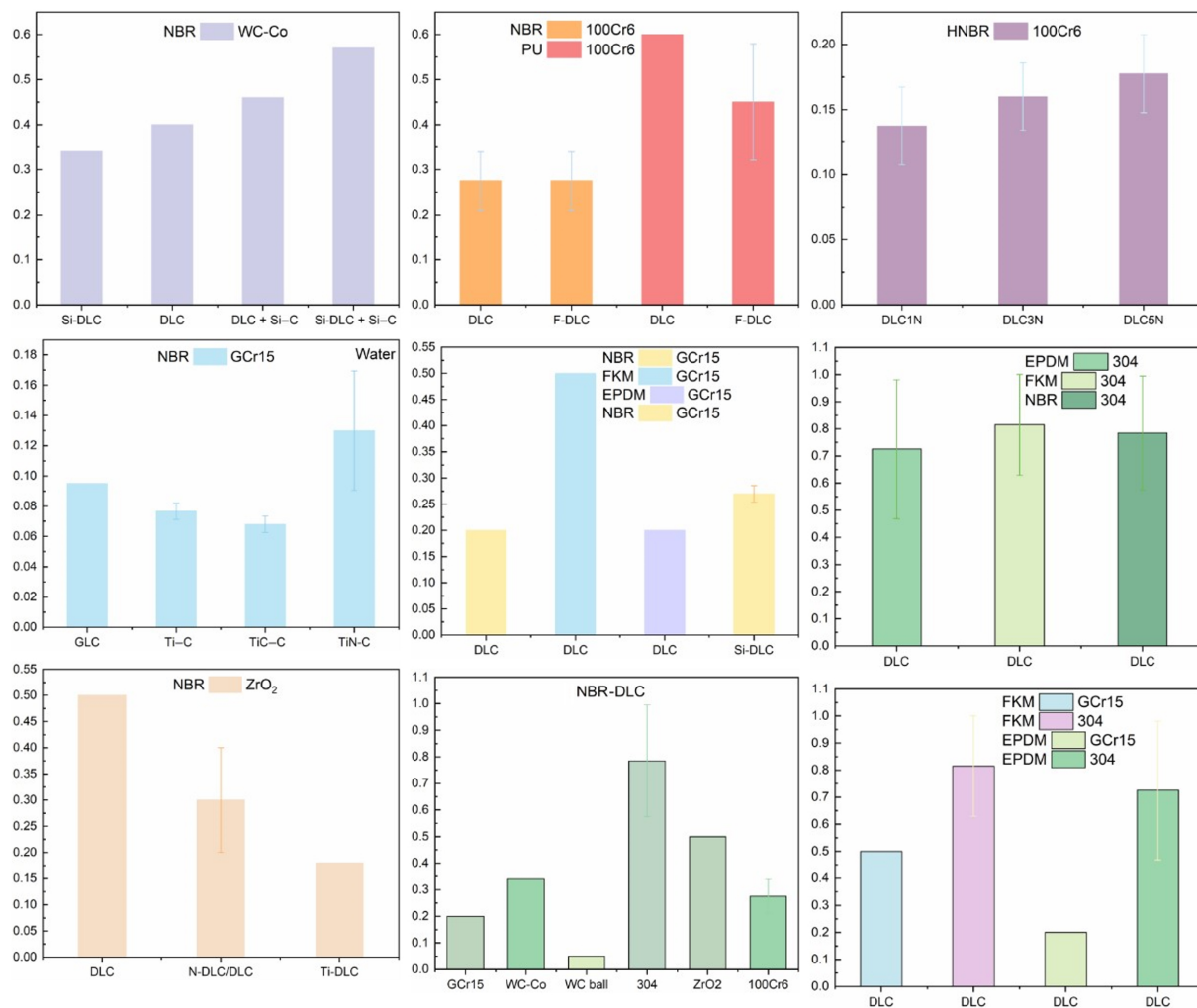


Fig. 4 Friction coefficient of the DLC film on the rubber surface.

strong material support for rubber seals and other tribological applications.

3.1 Friction behavior of carbon films on rubber surfaces

Carbon-based films have attracted widespread attention in the field of rubber surface modification because of their low friction, high hardness, and excellent wear resistance. The friction behavior of DLC films is influenced by multiple factors, including dopant elements, deposition processes, and friction conditions. Therefore, studying the tribological properties of DLC films on rubber surfaces, identifying their influencing factors, and developing optimization strategies are crucial for enhancing the service life and reliability of rubber sealing materials. As shown in Fig. 5, the friction coefficients of DLC films studied by different researchers in recent years are summarized.

3.1.1 Influence of dopant elements on friction behavior

To further enhance the tribological performance of DLC films, researchers have explored the doping of various elements, including Si, N, Ti, and F, into the DLC matrix. This doping process aims to improve the hardness, lubricity, adhesion, and wear resistance of the films. The incorporation of dopant elements can modify the chemical composition, sp²/sp³ ratio, and surface energy of the films, subsequently affecting their friction behavior.

A study by Lubwama et al. [167] in 2012 revealed that the friction coefficient of DLC and Si-DLC films increases with

friction distance, a phenomenon closely related to the microhardness of the films and the distribution of sp²/sp³ bonds. A higher microhardness helps maintain a lower friction coefficient, whereas the introduction of a Si-C interlayer reduces the wear resistance and stability of the films. These findings provide crucial tribological insights for the application of DLC films on rubber surfaces. In the same year, Lubwama et al. further demonstrated that the tribological performance of DLC and Si-DLC films on nitrile rubber surfaces is dependent on frictional conditions and silicon content. The DLC films presented lower friction coefficients and wear depths under dry sliding conditions, whereas the Si-DLC films presented superior tribological performance under wet sliding conditions. Additionally, water immersion significantly reduced the friction coefficient of the films, offering valuable tribological guidance for the practical application of DLC and Si-DLC films.

A study by Zhang et al. [176] in 2020 demonstrated that DLC and F-DLC films exhibited excellent tribological performance on NBR rubber substrates, significantly reducing the friction coefficient and providing outstanding wear resistance. However, for PU rubber substrates, fluorine doping may weaken film adhesion, leading to a higher friction coefficient and increased mass loss. These findings provide important theoretical insights for optimizing the application of DLC films on different rubber substrates (Fig. 6).

In 2022, Bai et al. [173] reported that DLC films with a hydrogen content of 29 at% exhibit optimal friction performance

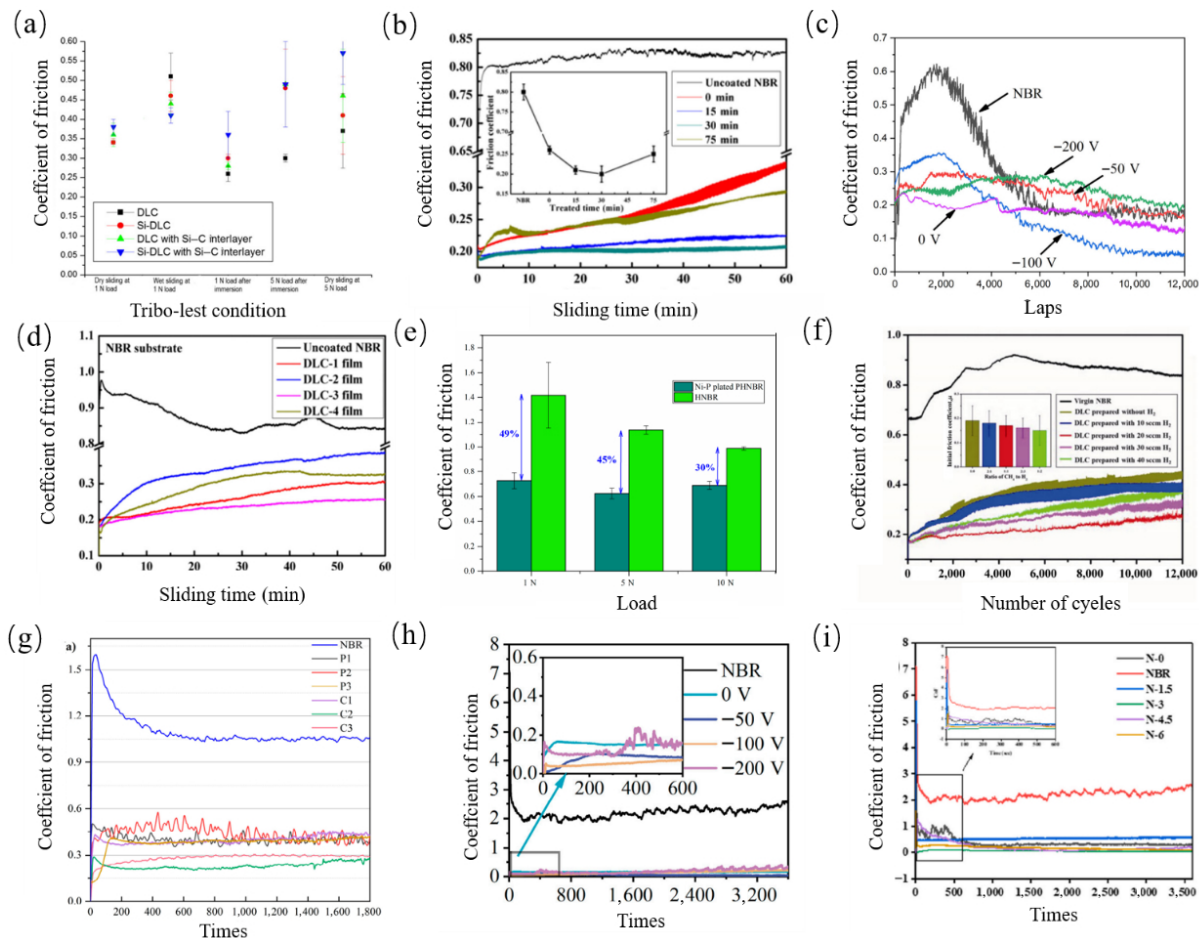


Fig. 5 (a) Average friction coefficient of DLC and Si-DLC coatings under different tribotesting conditions. (b) Friction coefficients of uncoated and coated NBR rubber samples with different pretreatment times. (c) Evolution of the coefficient of friction as a function of laps [178]. (d) Friction coefficient curves of films on NBR. (e) Mean and standard deviation of the CoF for the last 1,000 cycles of the pin-on-disc test for the pristine HNBR and Ni-P-plated PHNBRs for each load. (f) Friction curves of uncoated NBR and DLC-coated NBR with different ratios of CH_4 to H_2 . The insert shows the initial friction value. (g) Coefficient of friction of NBR coated with composite film. (h) Coefficient of friction of each sample [180]. (i) CoFs of NBR, DLC, and N-DLC films coated on NBR. Reproduced with permission from Ref. [166] for (a), © Elsevier B.V. 2012; Ref. [174] for (b), © Elsevier B.V. 2018; Ref. [165] for (d), © Elsevier B.V. 2019; Ref. [179] for (e), © the authors 2020; Ref. [173] for (f), © The Authors 2021; Ref. [171] for (g), © The Authors 2024; Ref. [181] for (i), © Elsevier Ltd. 2023.

($\mu \approx 0.2$) on NBR rubber, with a 74% reduction in the friction coefficient compared with that of uncoated NBR. The underlying mechanisms were as follows: (1) Hydrogen plasma etching reduced surface roughness while promoting the rearrangement of sp^2 carbon clusters, forming a lubricating graphite phase; (2) At an appropriate hydrogen content, the surface energy of the film matched the contact area, minimizing interfacial adhesion; and (3) excessive hydrogen content (e.g., 31 at%) led to a decrease in film hardness (19.8 GPa) and excessive aggregation of the graphite phase, causing adhesive wear and weakening the load-bearing capacity. Unlike earlier studies that relied solely on chemical doping or hard-phase reinforcement strategies, this work highlighted the crucial role of hydrogen in regulating the microstructure. Moreover, the thermodynamic instability of rubber substrates could accelerate the random nucleation of the graphite phase, underscoring the need to optimize the hydrogen content to balance lubrication and durability (Fig. 7).

In 2024, Yang et al. [180] investigated the tribological performance of nitrogen-doped DLC films on nitrile rubber surfaces and reported that the friction coefficient of nitrogen-doped DLC (N-DLC) films initially decreased but then increased with increasing nitrogen flow rate. At a nitrogen flow rate of $3 \text{ cm}^3/\text{min}$, the film exhibited the lowest friction coefficient of approximately 0.057. This was attributed primarily to the lowest

surface roughness, minimal surface energy, highest sp^3 -hybridized carbon content, and greatest hardness under these conditions. The reduction in surface roughness minimized the formation of hard particles during friction, thereby mitigating the plowing effect and wear caused by these particles. Under the optimal nitrogen flow rate ($3 \text{ cm}^3/\text{min}$), the film demonstrated high hardness, low surface roughness, low surface energy, and strong adhesion, achieving excellent tribological performance characterized by a low friction coefficient and low wear rate.

In the same year, Song et al. [171] investigated the effects of a nitrogen-doped DLC buffer layer on the mechanical and tribological properties of DLC films deposited on nitrile rubber substrates. These findings revealed a strong correlation between the friction coefficient and the presence of the buffer layer: the introduction of the N-DLC buffer layer significantly reduced the friction coefficient of the composite film (N-DLC/DLC). The N-DLC/DLC composite film prepared at a bias voltage of -100 V (C2 sample) exhibited the lowest and most stable friction coefficient (~ 0.20) during friction testing. In contrast, the friction coefficient of the pure DLC film without a buffer layer was approximately 0.5, whereas that of the uncoated NBR substrate was approximately 1.0. This indicated that the N-DLC buffer layer effectively reduced the friction coefficient by altering the chemical bonding state of the carbon atoms (Fig. 8). Additionally, in 2024,

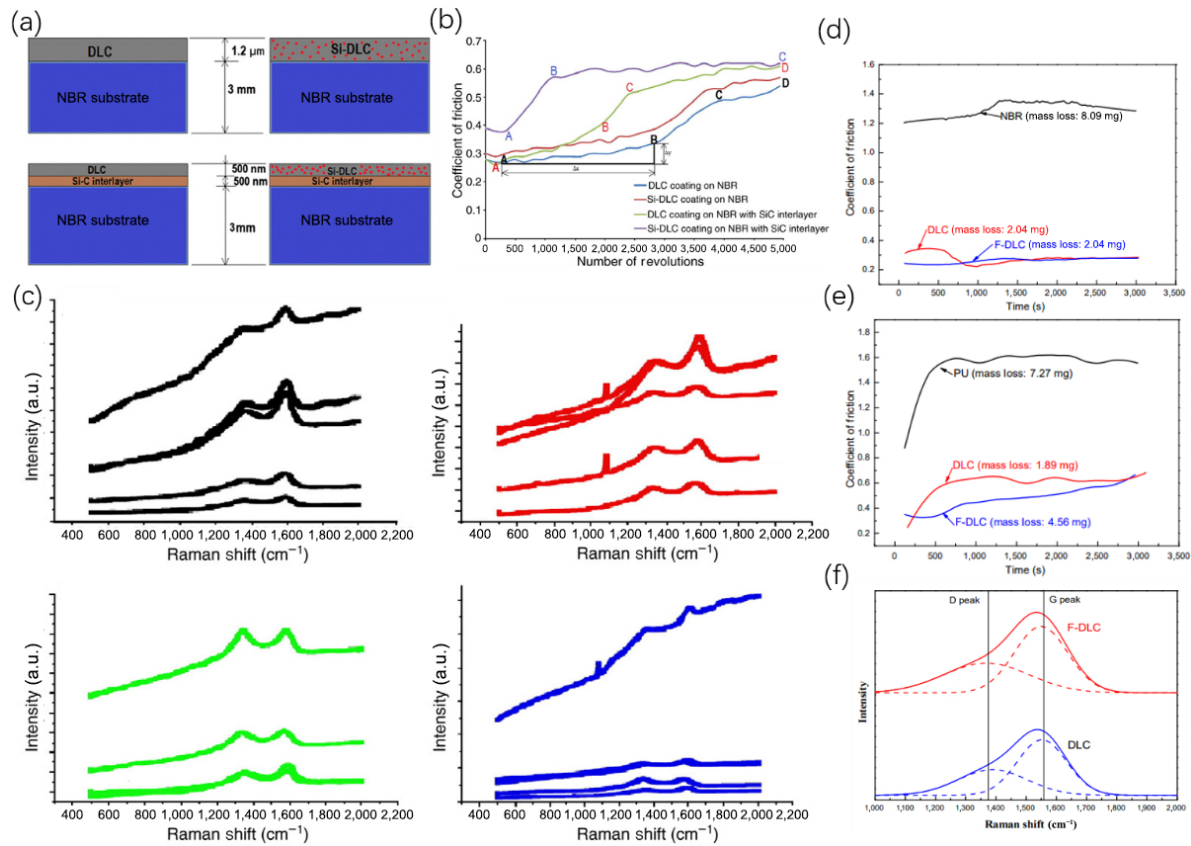


Fig. 6 (a) Process design for DLC and Si-DLC films deposited on an NBR substrate. (b) Coefficient friction vs. number of revolutions for DLC and Si-DLC with and without a Si-C interlayer. Points A, B, C, and D are indicated for estimation of slopes of the increase in the coefficient of friction. (c) Raman spectra for DLC, Si-DLC, DLC with Si-C interlayer and Si-DLC with Si-C interlayer films deposited on NBR substrate. (d) Coefficients of friction and film mass loss after tribotests of uncoated NBR, deposited DLC, and F-DLC films on ultra-high cis polybutadiene (UBR) substrates. (e) Uncoated PU rubber, deposited DLC and F-DLC films on PU rubber substrates. (f) Raman spectra of DLC and F-DLC films. Reproduced with permission from Ref. [167] for (a–c), © Elsevier B.V. 2012; Ref. [176] for (d–f), © The Authors 2020.

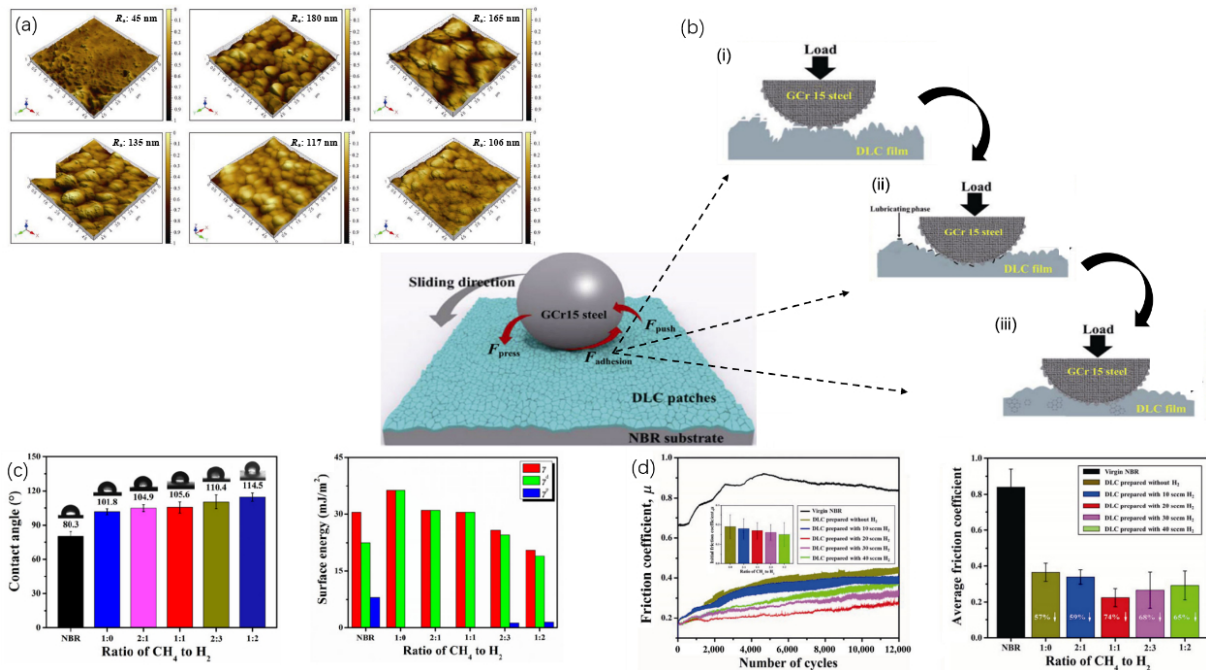


Fig. 7 (a) AFM images of uncoated NBR and DLC films deposited on NBR with different ratios of CH₄ and H₂ of 1/0, 2/1, 1/1, 2/3, and 1/2. (b) Schematic diagram of friction evolution between the counterpart and DLC films prepared with different hydrogen contents on rubber: (i) the low H-containing DLC film, (ii) the middle H-containing DLC film, and (iii) the high H-containing DLC film. (c) Steady-state CA values and images of a water drop. Total and component surface energies calculated via the OWRK method for uncoated and DLC-coated NBR with different ratios of CH₄ to H₂. (d) Friction curves of uncoated NBR and DLC-coated NBR with different ratios of CH₄ to H₂. The insert shows the initial friction values. Average friction coefficient of each group after three repetitions. Reproduced with permission from Ref. [173], © The Author(s) 2021.

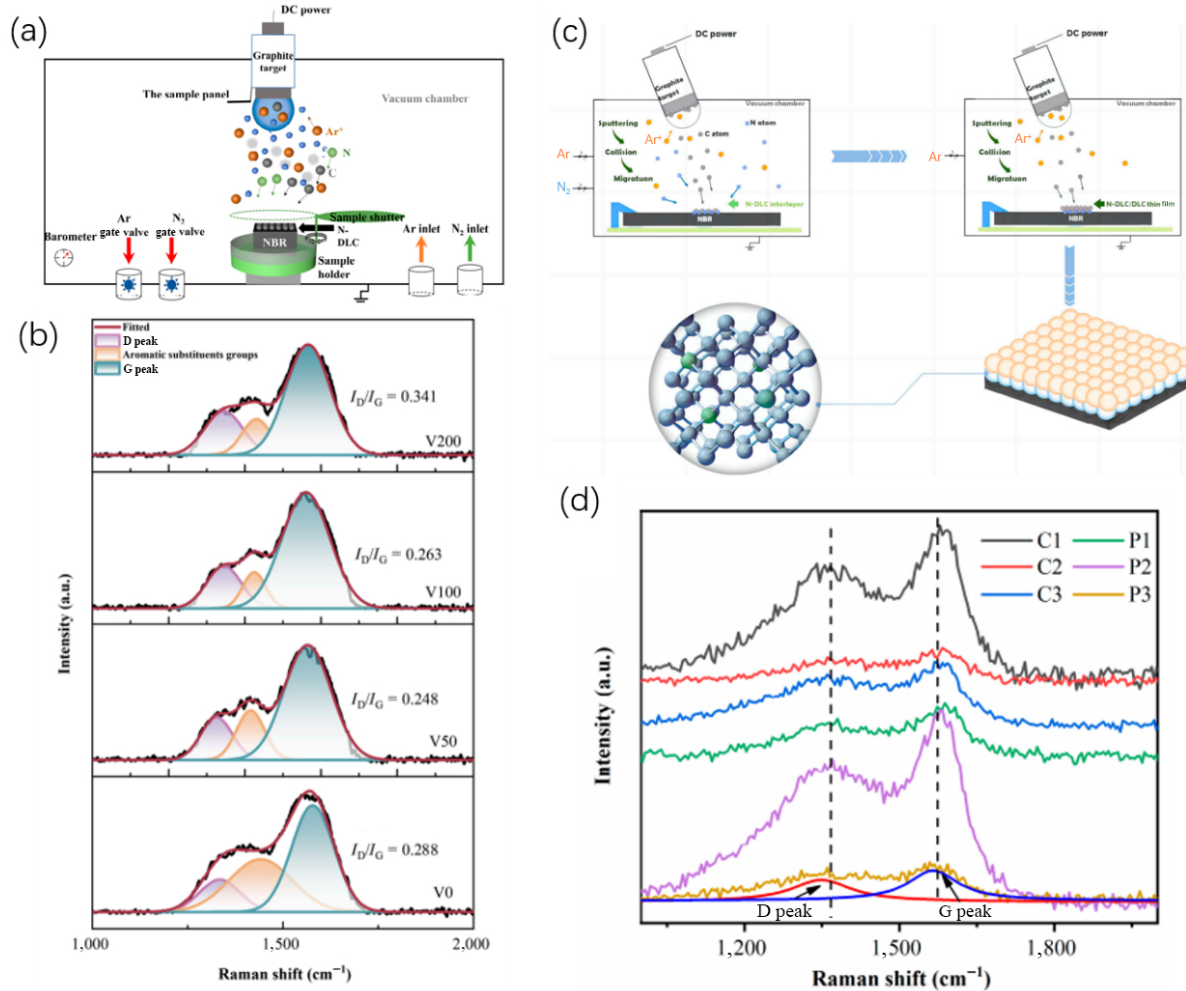


Fig. 8 (a) Schematic of the equipment used for the preparation of N-DLC films via magnetron sputtering. (b) Peak fitting of the Raman curves of each sample under different bias voltages to obtain the Raman curves of each sample and their respective I_D/I_G values. (c) Schematic diagram illustrating the deposition process of the N-DLC intermediate layer and the composite film. (d) Raman spectra of the DLC/N-DLC multilayer films and DLC films deposited on nitrile rubber under different interlayer bias voltages. Reproduced with permission from Ref. [180] for (a, b), © The Authors 2024; Ref. [171] for (c, d), © The Authors 2024.

Yang et al. [181] explored the tribological performance of N-DLC films on nitrile rubber substrates and examined the influence of bias voltage during DC magnetron sputtering on film properties. Tribological tests revealed that all the N-DLC films presented significantly lower friction coefficients than did the uncoated NBR. Among them, the V50 film had the lowest average friction coefficient (0.05) and demonstrated an extremely low wear rate ($\sim 10^{-9}$ magnitude) during friction testing. Wear track analysis revealed that the V50 film exhibited almost no visible wear marks, indicating excellent wear resistance. In contrast, the films deposited at bias voltages of 0 and -200 V presented higher wear rates and visible wear marks.

3.1.2 Influence of the deposition process on friction behavior

The tribological performance of DLC films depends not only on their chemical composition but also on their deposition process and process parameters. Different deposition methods result in significant variations in the microstructure, sp^2/sp^3 ratio, internal stress, and surface morphology of DLC films, which in turn affect their friction behavior. Additionally, deposition parameters (such as bias voltage, deposition temperature, and gas flow rate) influence the density, thickness, and surface energy of the film, thereby affecting its wear resistance and stability. Therefore, optimizing the deposition process parameters to achieve low

friction and high wear resistance is a key research direction for enhancing the tribological performance of DLC films.

In 1999, Tashlykov et al. [182] significantly improved the tribological performance of rubber by depositing a metal layer on its surface via a self-ion-assisted deposition technique. The carbon and oxygen components within the coating, along with the quasiperiodic surface structure, played a crucial role in reducing friction. The influence of different metal coatings on friction performance indicated that the reduction in friction force depended not only on the type of metal but also on the coating composition and deposition conditions. By optimizing these parameters, the tribological properties of rubber can be further enhanced, enabling superior performance in industrial applications.

A study by Pei et al. [163] in 2013 demonstrated that the segment size of DLC films can be adjusted by controlling the temperature variations of the rubber substrate during the deposition process, thereby significantly improving their tribological performance. DLC films with smaller segment sizes exhibited lower friction coefficients and superior wear resistance, providing essential design guidelines for depositing low-friction, highly wear-resistant DLC films on dynamic rubber sealing surfaces (Fig. 9).

A study by Bai et al. [174] in 2018 demonstrated that by

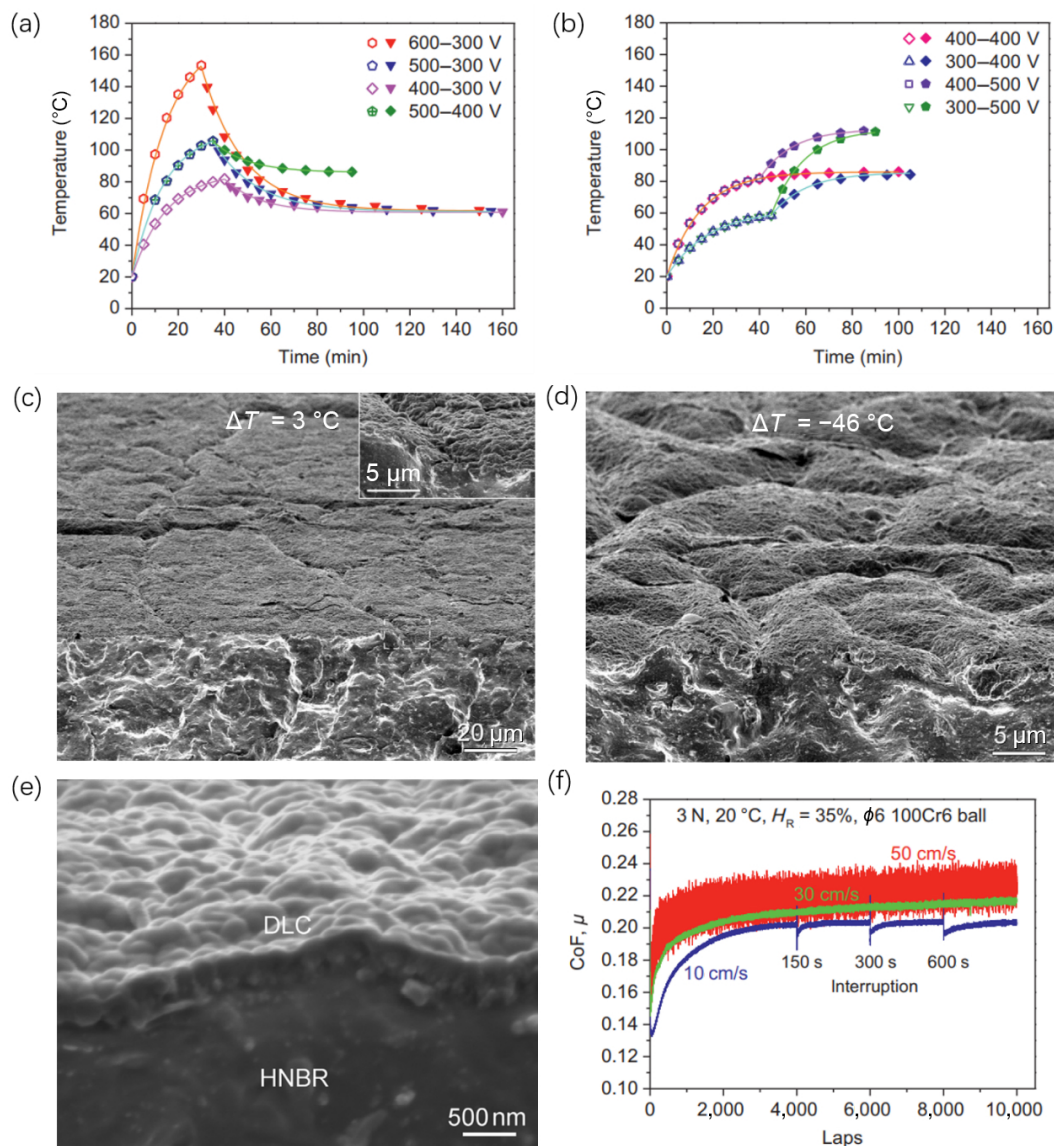


Fig. 9 (a) Temperature drop (negative T) and (b) temperature increase (positive T) during deposition. (c) Flat segments formed under tensile stress ($T = 3^\circ\text{C}$). (d) Convexly curved segments deposited under compressive stress ($T = -46^\circ\text{C}$). (e) Close-up view of a typical fracture cross-section of the DLC film-coated HNBR. (f) Effect of sliding velocity and interruption time on the friction of the DLC film-coated HNBR. Reproduced with permission from Ref. [163], © Elsevier B.V. 2013.

optimizing the argon plasma pretreatment time (30 min), DLC films with strong adhesion, low friction coefficients, and excellent wear resistance could be successfully deposited on NBR rubber surfaces. These films exhibited prolonged service life under high loads, significantly reducing the friction coefficient and wear rate of rubber. This provides an effective surface modification solution for rubber seals.

A study by Liu et al. [178] in 2019 demonstrated that by adjusting the bias voltage, DLC films with excellent tribological and sealing performance could be deposited on NBR surfaces. When the bias voltage was set at -200 V , the DLC-coated NBR sample presented the lowest friction coefficient, wear rate, and leakage rate. This was attributed to the higher hardness, lower sp^2/sp^3 ratio, and optimized surface roughness and Young's modulus. These findings provide crucial theoretical and experimental foundations for designing high-performance rubber seals. In the same year, Qiang et al. [168] reported that by employing high-bias ($-1,000\text{ V}$) argon plasma pretreatment, Si-DLC films with strong adhesion, low friction coefficients, and excellent wear resistance could be deposited on NBR surfaces.

Under high loads, these films presented the lowest friction coefficient and wear rate, significantly enhancing the tribological performance of NBR. This study offers valuable theoretical and experimental insights for the development of high-performance rubber seals.

A study by Vasconcelos et al. [179] in 2020 reported that Ni-P coatings deposited on HNBR surfaces via an electroless plating technique significantly reduced the friction coefficient of rubber and exhibited stable tribological performance under varying loads. The Ni-P coating demonstrated strong adhesion, excellent wear resistance, and insensitivity to load variations, making it a promising alternative to DLC coatings, particularly for dynamic sealing applications requiring high wear resistance and low friction coefficients.

4 Mechanical properties of the carbon films on rubber surfaces

The mechanical properties of carbide films on rubber surfaces primarily include hardness, elastic modulus (E), and adhesion

strength. However, the viscoelastic nature of rubber poses significant challenges in accurately measuring the mechanical properties of surface films.

4.1 Hardness and elastic modulus

In a tribological system, the hardness of a thin film determines its resistance to plastic deformation. Higher hardness (HIT) contributes to improved wear resistance; however, excessive hardness may increase brittleness, leading to crack propagation and delamination. Additionally, the elastic modulus influences the contact behavior of the film—a higher elastic modulus (EIT) can result in greater contact stress, whereas a moderate elastic modulus helps optimize the stress distribution and enhance frictional stability. Moreover, the adhesion strength between the film and the substrate is crucial, as a low adhesion strength may cause coating delamination during friction, compromising durability.

In recent years, researchers have optimized the mechanical properties of thin films using different dopant elements (such as nitrogen and titanium) and multilayer composite structures (such as DLC/Ti-C and DLC/N-DLC) to enhance their tribological performance. For example, N-DLCs can reduce internal stress and improve adhesion strength, thereby enhancing wear resistance. Titanium-doped DLC (Ti-DLCs) films can form TiC nanocomposite phases, increasing toughness and optimizing the friction coefficient. Additionally, substrate pretreatment and interface layer design are key strategies for improving the adhesion strength.

Owing to the elastic deformation of the rubber substrate, evaluating the hardness and elastic modulus of surface films remains challenging. Most researchers have employed nanoindentation testing to assess these mechanical properties. Table 5 summarizes the hardness and elastic modulus values of various films deposited on rubber surfaces, whereas Fig. 10 provides a clear visualization of the hardness and modulus data for carbon films on rubber substrates.

Hardness is a key factor in determining the wear resistance of thin films. A higher hardness enhances a film's resistance to plastic deformation, reducing material transfer or surface damage caused by contact pressure, thereby lowering the friction coefficient and wear rate. However, excessive hardness may also increase the brittleness of the film, making it more prone to crack propagation or delamination under high loads or high-speed sliding conditions. Studies have shown that optimal wear-resistant films require a balance between hardness and toughness to achieve a synergistic effect of low friction and high durability. The elastic modulus determines the deformation capacity of the film during contact. A higher E value enhances the film's load-bearing capacity, maintaining shape stability under high loads. However, it can also increase the surface contact stress, increasing the susceptibility of the film to localized cracking or delamination. Conversely, a lower E value provides better flexibility, reducing the risk of brittle failure, but may lead to a higher wear rate. Therefore, selecting an appropriate E value is crucial for optimizing tribological performance.

Among various hard films, studies have shown that doping with various elements (such as nitrogen, titanium, or silicon) can effectively regulate the sp^3/sp^2 bonding ratio in DLC, thereby influencing its hardness and tribological properties. For example, nitrogen doping can reduce internal stress and improve toughness, whereas titanium doping can form TiC nanocomposite phases within a carbon-based structure, enhancing wear resistance. Additionally, introducing an intermediate transition layer (such as N-DLC or Ti-C) can help

modulate the elastic modulus and improve the overall wear resistance of the film. For example, an N-DLC layer can reduce the E value of DLC films, increasing their compatibility with flexible substrates such as rubber and thereby increasing their overall durability.

Qiang et al. [183] demonstrated that the presence of a silicon (Si) interlayer significantly influences the growth mechanism and microstructure of a film. Although the Si interlayer did not markedly enhance the adhesion between the film and the NBR substrate, films containing the Si interlayer presented lower friction coefficients and wear rates under high bias voltage conditions. This indicates that the Si interlayer plays a crucial role in improving the load-bearing capacity and tribological performance of the film. Pei et al. [185] measured the hardness

Table 5 Hardness and elastic modulus of films on rubber surfaces

Substrate	Film	HIT (GPa)	EIT (GPa)	Ref.
NBR	Uncoated	2.9	15.2	[183]
NBR	DLC	5.5	39.6	[183]
NBR	Si-DLC	5	39.6	[183]
NBR	DLC/Si-C	5.1	39.6	[183]
NBR	DLC	3.6	39.6	[184]
NBR	Si-DLC	3.6	39.6	[184]
NBR	C30A	5.5	72	[185]
NBR	C45A	6.4	45.5	[185]
NBR	C60A	7.8	55.3	[185]
NBR	C75A	8.9	64.4	[185]
NBR	5Ti	0.0192	0.072	[186]
NBR	12Ti	0.009	0.0455	[186]
NBR	13.5Ti	0.0098	0.0506	[186]
NBR	15Ti	0.00783	0.04283	[186]
NBR	32Ti	0.0068	0.0196	[186]
NBR	N-0	0.0192	0.072	[181]
NBR	N-1.5	0.0192	0.072	[181]
NBR	N-3	0.033	0.072	[181]
NBR	N-4.5	0.0192	0.072	[181]
NBR	N-6	0.0192	0.072	[181]
NBR	DLC1	28.84±1	164.42±3	[174]
NBR	DLC2 (15 min)	27.59±3	142.35±10	[174]
NBR	DLC3 (30 min)	23.37±1	155.94±3	[174]
NBR	DLC4 (75 min)	21.57±1	134.21±3	[174]
NBR	DLC	15.5	—	[167]
NBR	DLC + Si-C	9.1	—	[167]
NBR	Si-DLC	11.1	—	[167]
NBR	Si-DLC + Si-C	12.1	—	[167]
NBR	Cr-DLC	60–80		[177]
FKM	Cr-DLC	60–80		[177]
EPDM	Cr-DLC	60–80		[177]
EPDM		60–80	6.65–16.53	[169]
FPM		60–80	3.44–16.06	[169]
NBR		60–80	3.41–16.75	[169]
NBR	Ti/N-GLC	7.9–12.9	120.9–144.4	[170]
NBR	Mo-Se-C	7.4	105	[187]
NBR	N-DLC	6.8		[180]
NBR	H-24%	24.8±0.5	145.5±5	[173]
NBR	H-26%	23.2±0.8	140.3±2	[173]
NBR	H-29%	21.8±0.6	134.3±2	[173]
NBR	H-30%	20.9±0.6	128.6±5	[173]
NBR	H-31%	19.8±0.9	129.6±4	[173]

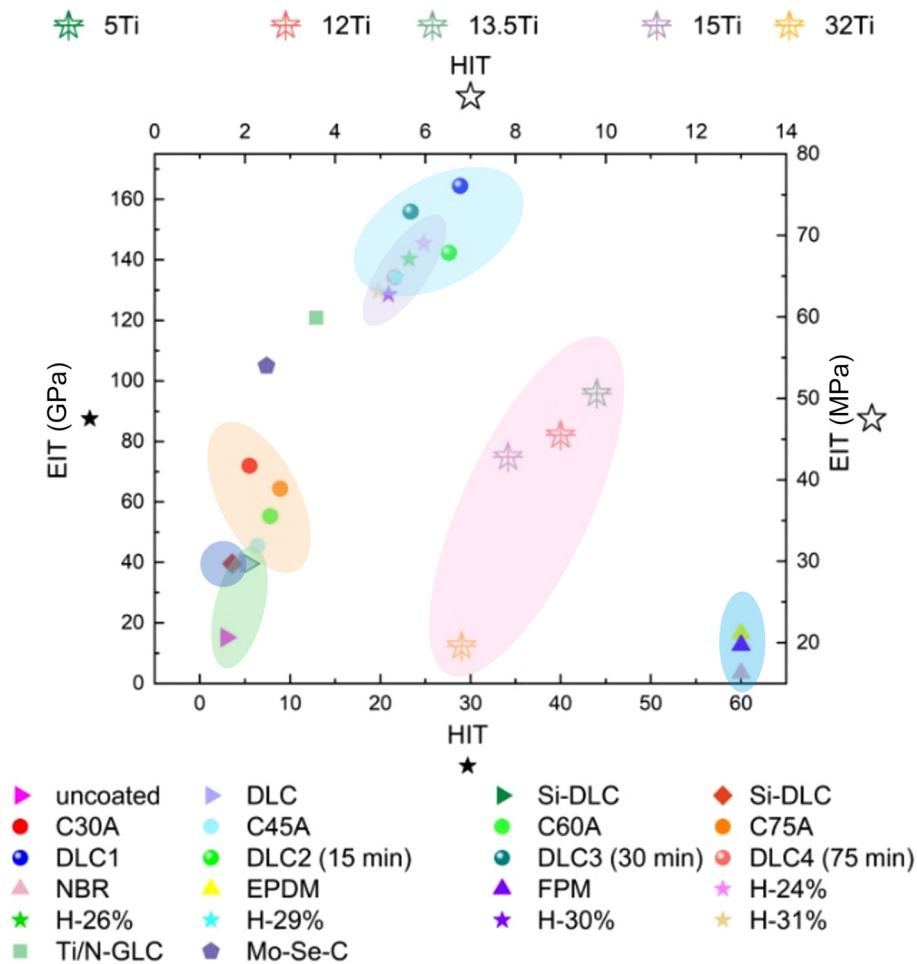


Fig. 10 Comparison of the hardness and elastic modulus of the carbon films on different rubber surfaces.

and elastic modulus of a-C:H films and reported that as the arc current increased, both the hardness and elastic modulus improved, along with a significant increase in the film adhesion strength. However, excessive arc current led to increased surface roughness and reduced flexibility, negatively impacting the tribological performance. Their study revealed that the hardness and elastic modulus of a-C:H films are closely related to their microstructure and hydrogen content. An optimal hydrogen concentration was shown to increase hardness and wear resistance while maintaining good flexibility and adhesion. Furthermore, Yang et al. [181] used nanoindentation tests to measure the hardness and elastic modulus of nitrogen-doped DLC films. The results indicated that nitrogen doping significantly increased both the hardness and elastic modulus while reducing the surface roughness and surface energy, thereby improving the film's tribological performance. At a nitrogen flow rate of 3 sccm, the N-DLC film presented the lowest friction coefficient (~ 0.057) and wear rate (on the order of 10^{-9}), demonstrating outstanding tribological performance. This was attributed to the high hardness, strong adhesion to the substrate, low surface roughness, and reduced interfacial adhesive forces of the film (Fig. 11).

4.2 Interfacial adhesion mechanism

The interfacial adhesion mechanism involves both physical and chemical interactions. Plasma pretreatment significantly enhances the adhesion strength by effectively cleaning the surface, promoting chemical bonding, and optimizing the surface energy and wettability. The design of transition layers further improves adhesion performance by alleviating stress, enhancing chemical

compatibility, and optimizing the interfacial energy. The experimental results indicate that argon plasma pretreatment and silicon interlayers substantially enhance the adhesion strength between DLC films and rubber substrates.

4.2.1 Evaluation of the interfacial adhesion mechanism

The adhesion strength between the film and the rubber substrate is a critical parameter for evaluating the surface performance of coated rubber. Owing to the elastic nature of rubber substrates, conventional adhesion measurement methods are often limited. Currently, researchers commonly use the tape peel test to assess the adhesion strength of DLC films on rubber surfaces.

In this method, an X-cut is made on the coated rubber surface, followed by the application of semitransparent pressure-sensitive adhesive tape, which is then quickly peeled off. The extent of film detachment at the incision site was examined under a microscope to qualitatively evaluate the adhesion strength of the film [167, 188]. This technique provides an effective way to estimate film adhesion, although it remains qualitative. To achieve more precise quantification, advanced methods such as scratch testing or nanoindentation-based adhesion analysis may be considered in future studies.

4.2.2 Effect of plasma pretreatment on interfacial adhesion

Radicals and dangling bonds generated by plasma treatment can form chemical bonds, such as covalent or hydrogen bonds, with carbon atoms in the film, thereby significantly enhancing interfacial adhesion. Qiang et al. [183] utilized argon plasma pretreatment, Masami et al. [189] employed ozone as the plasma

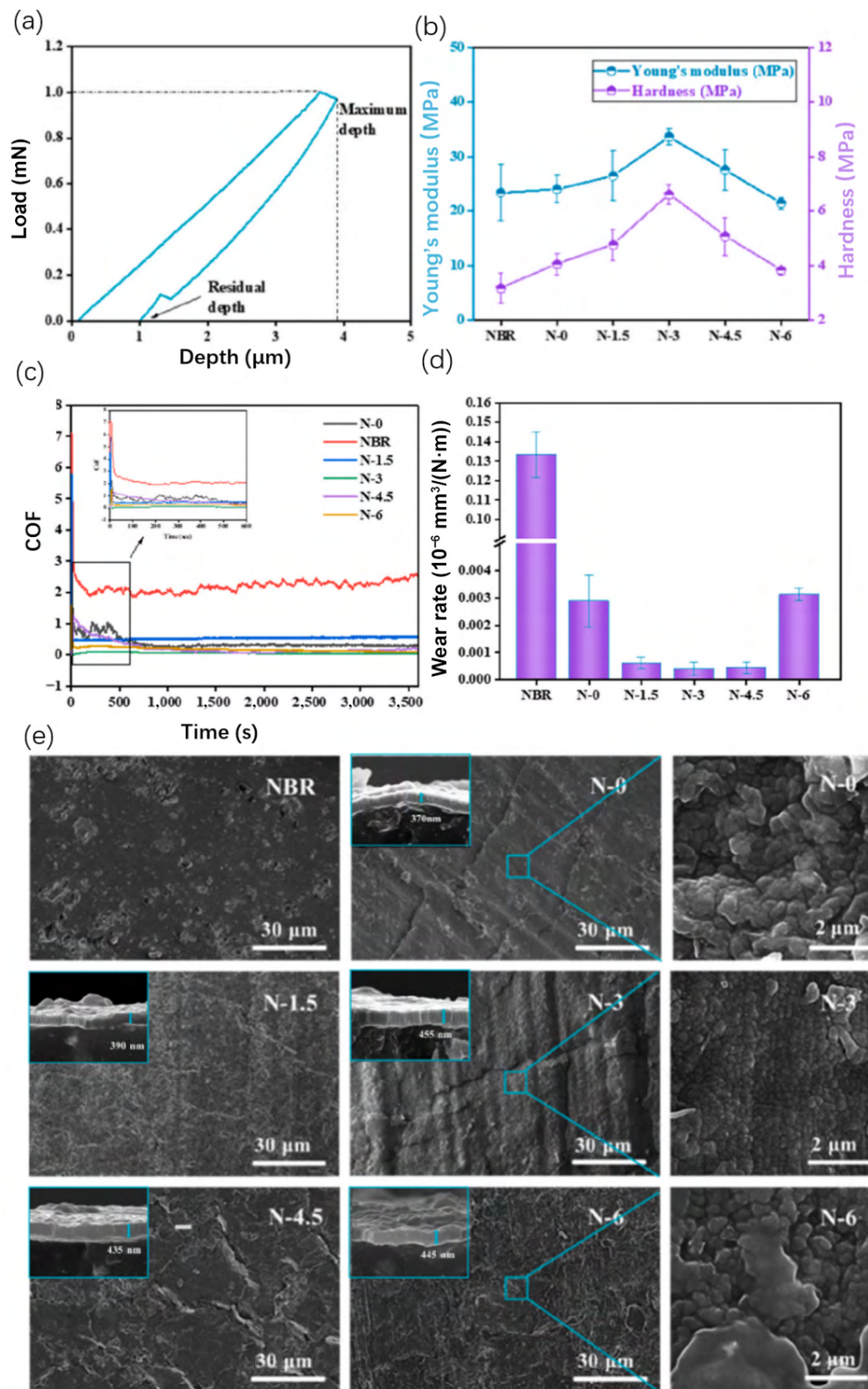


Fig. 11 (a) Load-depth curves of sample N-3 deposited on the NBR surface. (b) Hardness and Young's modulus variation curves of NBR and N-DLC films deposited on the NBR surface. (c) CoF and (d) wear rates of NBR, DLC, and N-DLC films coated on NBR. (e) Surface morphology of NBR and surface and cross-sectional morphology of DLC and N-DLC deposited on the NBR surface. Reproduced with permission from Ref. [181], © Elsevier Ltd. 2023.

source, Kim et al. [190] used O_2 plasma, and Lubwama et al. [166, 167, 184] adopted Ar as the plasma pretreatment gas.

Similarly, Guo and Hong [188] applied inert argon plasma for surface pretreatment. The effectiveness of argon plasma is

attributed to its ability to break C-H bonds on the rubber surface, generating dangling bonds that facilitate carbon atom bonding during film deposition, thereby improving the adhesion strength. Cleaning the rubber substrate to remove surface contaminants has been proven effective in improving the film adhesion strength [191, 192]. Following the cleaning process, plasma pretreatment of the rubber surface further enhances the adhesion strength of the film.

Experimental results from Bai et al. [174] revealed that untreated DLC films exhibited significant delamination when subjected to tape peeling, whereas DLC films pretreated with Ar plasma showed substantially reduced delamination, indicating increased adhesion strength. Similarly, Qiang et al. [168] reported that untreated Si-DLC films experienced noticeable delamination during tape peeling, whereas those subjected to argon ion pretreatment exhibited less delamination, further confirming

improved adhesion strength. This enhancement is attributed to the argon ion treatment, which effectively cleans the NBR surface and generates free radicals, thereby strengthening the chemical bonding between the film and the NBR substrate (Fig. 12).

4.2.3 Effects of transition layer design on interfacial adhesion

The transition layer, which serves as a load-bearing intermediary, can significantly enhance the adhesion strength between the film and the substrate. Its mechanisms include alleviating stress and utilizing chemical compatibility.

Alleviating stress: The transition layer can effectively relieve the compressive stress of the film, thereby reducing delamination. For example, in 2008, Pei et al. [193] investigated the use of a Cr interlayer and reported that the presence of a Cr interlayer was detrimental to the film-substrate adhesion strength. In 2019, Qiang et al. [165] employed a Si interlayer and experimentally

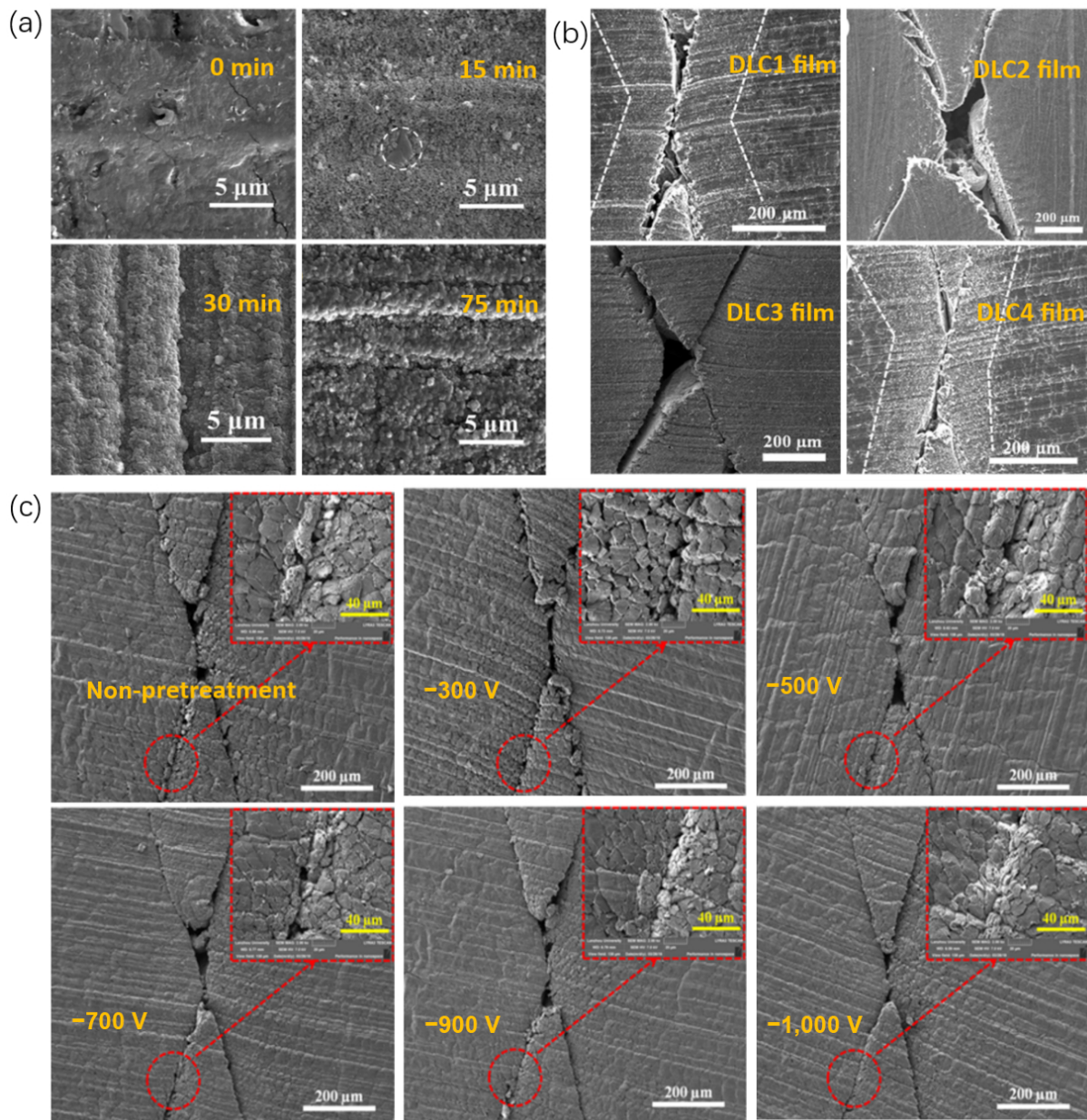


Fig. 12 (a) Surface morphology of NBR rubber pretreated with Ar plasma at different times of 0, 15, 30, and 75 min. (b) SEM images of X-cut locations after peel tests for DLC1 film, DLC2 film, DLC3 film, and DLC4 film. (c) SEM images of X-cut locations after peel tests for the film deposited at different pretreatment biases of nonpretreatment: -300, -500, -700, -900, and -1000 V. Reproduced with permission from Ref. [174] for (a, b), © Elsevier B.V. 2018; Ref. [168] for (c), © The Author(s) 2019.

determined that at an optimal Si interlayer thickness of 1.04 μm , the Si-DLC film exhibited superior adhesion strength with minimal delamination. However, at other thicknesses, delamination was more pronounced. This effect is attributed to the optimal Si interlayer thickness, which effectively relieves the compressive stress of the film and enhances its compatibility with NBR. Chemical compatibility: The selection of transition layer materials is crucial for interfacial adhesion. In 2019, Wu et al. [172] introduced Ti-doped carbon films as an interlayer, demonstrating that appropriately biased interlayer deposition

significantly enhanced the adhesion strength (Fig. 13(a)). Specifically, Ti forms strong Ti-C bonds with dangling carbon atoms on the surface of the rubber substrate, whereas the formation of TiC nanocrystals further enhances the interfacial bonding. Additionally, the Ti-C interlayer reduces the residual stress in the DLC film and mitigates delamination (Figs. 13(b)–13(e)).

The hardness, elastic modulus, and adhesion strength of a thin film do not independently affect its tribological performance; rather, they collectively affect its wear resistance. Optimizing the

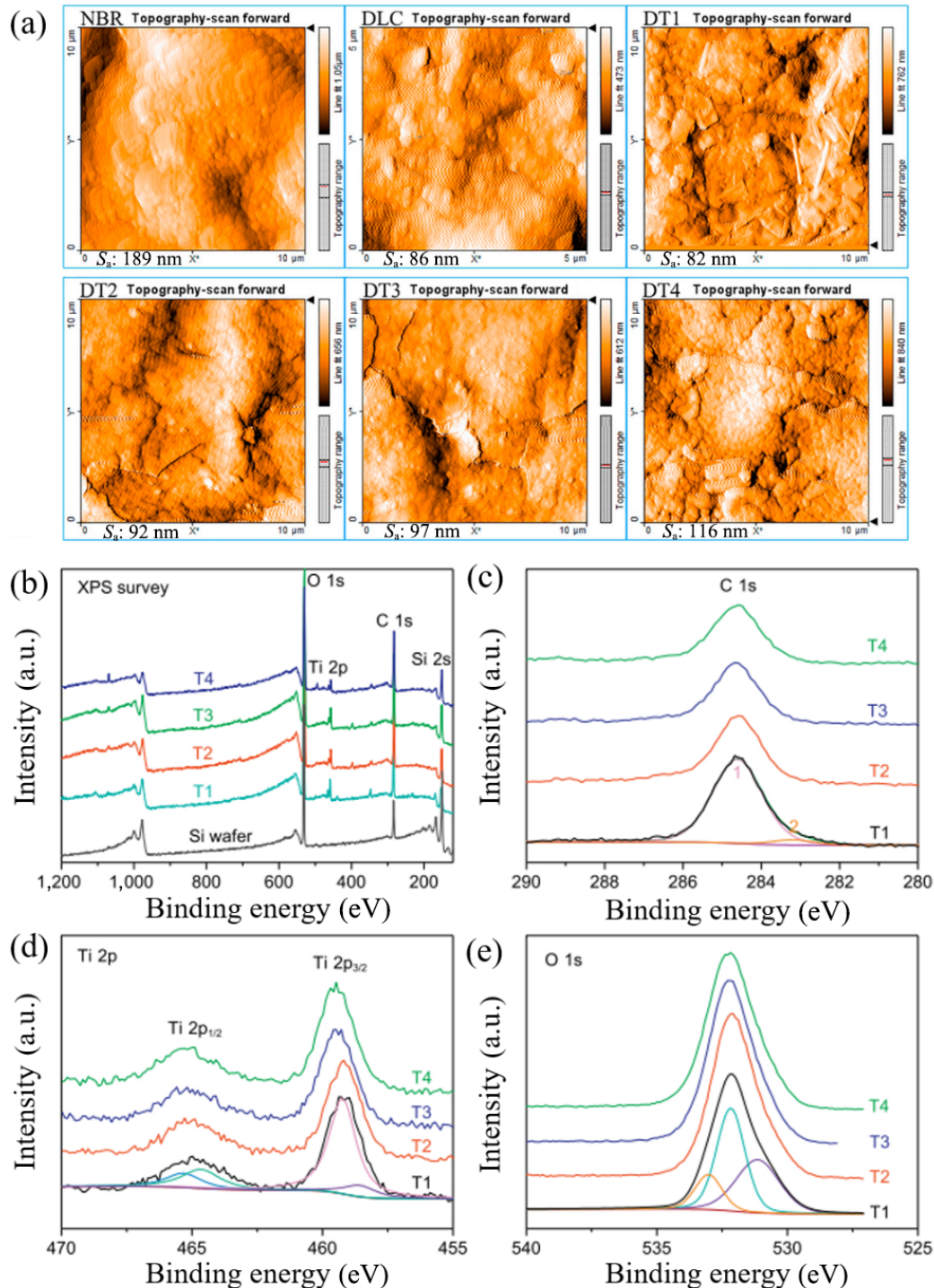


Fig. 13 (a) AFM images with a scan area of 10 $\mu\text{m} \times 10 \mu\text{m}$ of NBR, DLC, DT1, DT2, DT3, and DT4 films coated on NBR. XPS survey spectra and high-resolution XPS scan spectra. (b) XPS survey spectra and high-resolution XPS scan spectra over (c) C 1s, (d) Ti 2p, and (e) O 1s of the Ti-C layer deposited on Si wafers under different substrate negative bias voltages (0, -50, -100, and -150 V, marked as T1, T2, T3, and T4, respectively).

synergistic effect of these mechanical properties can effectively enhance the film's tribological behavior. For example, adjusting the sp^2/sp^3 carbon bond ratio, introducing dopant elements, or optimizing multilayer structures can improve wear resistance while maintaining a low friction coefficient. Future research can further integrate experimental measurements with numerical simulations to explore more precise tribological optimization strategies.

5 Summary and outlook

This paper systematically reviews the research progress on the use of carbon-based films on rubber surfaces for enhancing the tribological properties of rubber. As described in Fig. 14, this work provides a detailed summary of various film materials, preparation methods, friction performance, and mechanical properties, including hardness, elastic modulus, and adhesion strength. The following key points emerge:

(1) Significant improvements in friction performance are observed with the deposition of DLC and its modified films on rubber surfaces. This process typically leads to a noticeable reduction in the friction coefficient and a substantial increase in the wear resistance. These benefits can be attributed primarily to the low friction characteristics of the films, their high hardness, and various mechanisms that enhance interface lubrication through the control of their crystal structures and other methods.

(2) Optimization of the preparation process and film structure: Different deposition techniques (such as magnetron sputtering, PLD, and PECVD) and deposition parameters (such as bias voltage, argon plasma treatment, and the introduction of doped elements) significantly affect the microstructure and properties of

the films. Proper process optimization not only improves the hardness and adhesion strength of the films but also enables control of the sp^2/sp^3 bond ratio, resulting in a lower friction coefficient and enhanced wear resistance.

(3) Interface adhesion and compatibility issues: Due to the soft, viscoelastic nature of the rubber substrate, there are interface stress and adhesion mismatch issues between the rubber and the hard film. Surface pretreatment and the introduction of intermediate layers (such as silicon layers or Ti doping) can improve the adhesion strength between the film and the rubber, laying the foundation for practical applications.

Although significant progress has been made in improving the tribological properties of rubber, several unresolved issues and future research directions remain:

(1) In-depth study of interface interaction mechanisms: The chemical bonding, interface stress distribution, and thermal mismatch effects between rubber and carbon-based films still require further quantitative research. Combining experimental characterization with theoretical simulations to explore the interaction mechanisms between films and soft substrates will help guide the optimization of preparation processes.

(2) Fine-tuning of film microstructures: Future research can focus more on controlling the sp^2/sp^3 ratio, the type and content of doped elements, etc., to achieve an ideal film structure that meets both low friction requirements and balances wear resistance and flexibility.

(3) Development of new multifunctional composite films: With the continuous advancement of materials science and artificial intelligence technologies, combining different types of solid lubricants (such as metals, ceramics, and carbon-based materials)

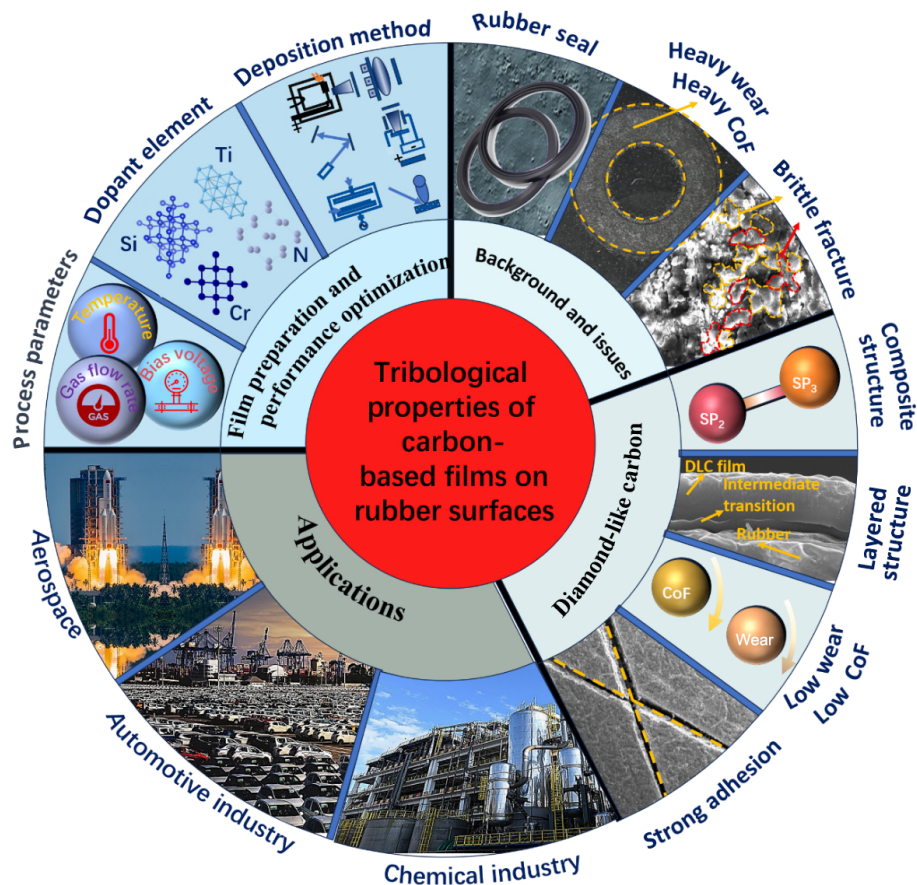


Fig. 14 A review diagram of the tribology of carbon-based films on rubber surfaces.

to develop rubber surface modification films with multifunctional properties, such as self-healing, antipollution, or smart responsiveness, will be an important research direction in the future.

(4) Industrial application and large-scale fabrication technologies: Many high-performance film preparation techniques have been validated at the laboratory stage. However, the key to their widespread application lies in transforming these technologies into cost-effective, stable, and scalable industrial processes suitable for large-area continuous production. Future research will need to focus on process equipment, deposition rates, and environmental adaptability.

In conclusion, carbon-based film modification technology for rubber surfaces has significant potential for improving tribological properties. Future research should not only focus on breakthroughs in fundamental mechanisms but also address engineering issues related to practical applications. Through interdisciplinary collaboration, there is hope for technological transformation and widespread application in high-performance rubber seals, dynamic friction systems, and other related fields.

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Declaration of competing interest

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

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Weiye Li obtained his bachelor degree from Shenyang University of Chemical Technology, China, in 2019. Currently, he is pursuing a master degree at Lanzhou University of Technology. His current main research interests include the growth of nanostructured films and their tribological properties, as well as the surface synergistic lubrication of solid films.



Li Chen is a professor in the School of Petrochemical Technology at Lanzhou University of Technology, China. She received her Ph.D. degree in physical chemistry from Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, China, in 2010. Her research interests include the design and construction of lubricating films and the corrosion, wear and protection of engineering materials.



Bin Zhang received his Ph.D. degree in July 2011 from the Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, China, and received his bachelor degree in July 2005 from Lanzhou University, China. Currently, he is working at the Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, China, as a professor. He is a Fellow of the International Association of Advanced Materials (IAAM). He has published more than 116 papers on small, nano energy, carbon, friction, etc., and granted 58 patents authored via China, USA, and Japan. He has authorized the second prize of National Technological Invention (2016), the second prize of the National Scientific and Technological Progress Award (2019), and 4 other province departmental level awards.