Multilayer Coatings for Tribology

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Friction and wear usually lead to huge energy loss and failure of machine pairs, which usually causes great economic losses. Researchers have made great efforts to reduce energy dissipation and enhance durability through advanced lubrication technologies. Single-layer coatings have been applied in many sectors of engineering, but the performance of single-layer coatings still has many limitations. One solution to overcome these limitations is to use a multilayer coating that combines different components with varied physical and chemical properties. In addition, multilayer coating with alternating layers only containing two components can lead to improved performance compared to a coating with only two different layers.

multilayer coating friction wear tribology transition-metal nitride

diamond-like carbon

1. Introduction

Friction and wear occur in moving pairs with direct contact among all mechanical systems, leading to excessive energy consumption and failure of equipment [1]. Advanced techniques have been proposed to reduce friction and wear [2][3]. One of the methods is to deposit coating materials on friction pairs, which has been widely used for a long time due to its high performance in practical engineering applications. Coatings can be designed with different materials and structures to provide multiple functions. With the development of coating systems, different designing procedures have been proposed to further enhance the performance of coatings, or to extend the adaptivity of coatings in various environments [4][5][6]. One of the strategies is the multilayer designing of coating systems. Among the coating design concepts, multilayer coatings have attracted a lot of attention because the properties including hardness, elastic modulus, lubrication performance, and adhesion to substrate can be targeted and regulated, making it easier to develop coating systems to meet specific requirements.

2. Development of Multilayer TMN Coatings

The failure of tools can lead to great costs induced by the stopping of production and new adjustment of machines. Around 10% of the losses can be reduced through optimizing the lifetime of finishing parts [7]. Decreasing the wear of tools, better control of the forming process, and a reduction in lubricant and cleaning agents are the motivations to promote the development of coatings. Chemical vapor deposition (CVD) and physical vapor deposition (PVD) are commonly used techniques for the fabrication of coatings, where CVD coatings usually have superior properties compared to PVD coatings due to the higher deposition temperature. However, CVD coatings have higher cost and more time consumption, but limited friction-reduction performance [8]. Attributed to the high performance and relatively low cost, PVD coatings are a promising alternative of CVD coatings for wider industrial applications.

The transition-metal nitride (TMN) coatings are widely used in engineering applications due to their superior physical and chemical properties, including high hardness, wear resistance, thermal stability, and excellent corrosion and oxidation resistances [9][10][11][12]. TMN coatings are used as protective layers for cutting tools, molds, dies, or for abrasion and corrosion resistance in various fields, including aerospace, automotive, etc. The performances of several TMN coatings, including TiN [13][14][15][16], CrN [17][18][19][20], ZrN [21][22][23][24], MoN [25][26][27], NbN [28][29][30], and TaN [31][32], have been investigated for many years. The continuous development of advanced coating materials is motivated by the increased demands for industrial applications. It was showed that multilayer structure design can effectively improve the mechanical, chemical, as well as tribological properties of coatings [33]. In the multilayer coating, each layer exhibits a specific property, such as a thermal and diffusion barrier, adhesion to substrate, load carrying, lubrication, or wear resistance. The investigation of multilayer PVD coatings started in the 1970s [34][35] and was based on the models proposed by Koehler [36]. The model indicates that materials with high yield strength could be fabricated through alternating thin layers with different shear modules due to the inhibition of dislocation formation and mobility. The Al/Al_xO_y coatings were deposited, and the performances of the coatings were investigated. It was found that based on the layer spacing, a Hall–Petch-type relationship was obeyed for yield stress [34]. Bunshah et al. also studied both the metal/ceramic [37] and metal/metal [38] coatings deposited with evaporation techniques. In general, mechanical properties of the coatings can be improved with decreased layer thickness.

Holleck et al. fabricated multilayer coatings with different ceramic materials, demonstrating that the improvements in adhesion, indentation toughness, hardness, and wear-resistance performance can be achieved with optimized layer thickness [39]. The improved performance of multilayer coatings is believed to partially attribute to the stress relaxation and crack deflection, which exists in various contact conditions with cyclic loading and fatigue. Besides the influence of yield strength, the stacking sequence also has influence on the coating performance. It was found that the alternation of layers with high/low shear modulus can provide more benefits for multilayer DLC/metal carbide coatings [40] or TiN/Ti coatings [41][42].

To further investigate the influence of the stacking sequence of multilayer coatings, it is effective to consider the coatings under a point or distributed load, causing the deflection of coatings and the deformation of substrates [33]. Under such a circumstance, the maximum stress would increase with increased coating thickness if considering the bending stress. If the coatings are isolated as individuals to exclude the influence of substrate, each layer in multilayer coatings has much less stress compared to that of the thick layer if each layer can slide over each other. Hence, the alternating of hard/soft layers can offer a shear zone to prevent the fracture of hard and brittle layers under deflection induced by applied load. Moreover, there are also some other influences of the layer thickness. Since the layers also need to support the normal load, the minimum thickness of the soft layers is limited to provide adequate support to the hard layers. Additionally, the increased layer thickness will lead to increased relative sliding distance between each layer. Incorporating many thin layers is an ideal way to ensure the load-support properties. The layer thickness also depends on the loading condition and the aimed application of the multilayer coatings. For example, larger coating thickness is usually needed with the presence of hard and coarse third bodies. The benefits of the structure consisting of layers with relatively high hardness and relatively low hardness were also revealed via cyclic impact test [43][44]. The wear of coatings induced by plastic deformation can be suppressed through the composition of soft and hard layers, indicating that multilayer coatings have better prospect as a solution for various engineering problems. The concept and mechanisms of multilayer coatings with distinct performances were introduced [45][46]. Even a simple two-layer structure exhibited better wear- and corrosion-resistance properties [47][48]. The theoretical investigations have great value for better structural optimization of the multilayer coatings.

With the main elements of the coatings, TMN coatings can be divided into Ti-based TMN coatings, Cr-based TMN coatings, and TMN coatings with other elements. Ti and Cr are widely used as adhesion layer between the multilayer coating and metal substrates, which can be attributed to that the high binding energy makes it easier to form carbides, nitrides, and oxides on the adhesion layer. However, the performance of multilayer coatings strongly depends on the structure [49], which should be carefully decided for the designing of multilayer coatings. Usually, the multilayer coatings tend to achieve low friction, high wear resistance, good adhesion, and suppressed cracking during friction process.

3. Development of Multilayer DLC Coatings

Based on the concept of multilayer designing of coating systems, the multilayer ceramic metal-DLC coatings were fabricated by Voevodin et al. [45] using electron-enhanced unbalanced magnetron sputtering for sliding wear applications. Low COF and low wear rates can be achieved by the multilayer coatings with upper Ti20%-DLC and Ti35%-DLC layers. Sui et al. [50] prepared CrN/DLC/Cr-DLC multilayer coatings with plasma-enhanced chemical vapor deposition, which can significantly improve the lubrication and wear-resistance performance comparing to single component coating. The improved performances of the CrN/DLC/Cr-DLC multilayer coatings can be attributed to the lubrication of DLC layers, the supporting of CrN layers, the enhanced crack propagation inhibition, and the increased elastic recovery governed by the multilayer structure. DLC coatings were also combined with MoS_2 to enhance the lubrication and wear-resistance performances. Pu et al. [51] prepared a multilayer DLC/MoS_2 coating using medium-frequency magnetron sputtering, which exhibited a low COF of 0.02 and a low wear rate of ~6.5×10^{-6} mm^3 N^{-1} m^{-1}. The influence of different underlayers on the tribological behaviors of the DLC-based multilayer coatings prepared by magnetron sputtering was investigated by Duminica et al. [52], where a better adhesion could be achieved with only Cr under layer, exhibiting lower COF compared to other samples.

For single-layer DLC coating, high residual stress would lead to brittle fracture and delamination under high normal load during the friction process. Researchers found that the tribological behaviors of multilayer DLC coatings can be improved through the DLC layers with different properties. Li et al. [53] fabricated multilayer DLC coatings with alternated soft and hard layers through the alternating of bias during magnetron sputtering. Delamination was observed in monolayer coatings due to high residual stress. The results showed that the bonding structure (sp^3 and sp^2) can be changed by substrate bias. The sp^3 fraction in DLC coating can be increased with increased bias ratio on the two adjacent sublayers from −40 V/−160 V to −80 V/−160 V, leading to increased coating hardness. With the multilayer designing, the hardness of multilayer DLC coating was similar to the coatings deposited at low constant bias, but the adhesion strength and toughness were significantly improved. It can be concluded that alternately biased sputtering deposition is a promising way to fabricate DLC coating with high hardness, toughness, and adhesion strength. With the similar designing concept, Harigai et al. [54] fabricated multilayer N-DLC coatings with each layer thickness of 10 nm using filtered arc deposition, containing periodic bilayer structures with ta-C:N and soft a-C:N layers. The multilayer coatings showed better wear-resistance performance than monolayer ta-C:N coating and multilayer N-DLC coatings with each layer thickness of 50 nm. Lin et al. [55] fabricated multilayer DLC coatings with alternated soft and hard layers using unbalanced closed-field magnetron sputtering to enhance wear-resistance performance at high contact stress. It was found that the multilayer coating with a soft top layer had lower wear volume under high contact stress, which can be attributed to the fact that the soft top layer can form a transfer layer to reduce friction and wear.

4. Other Multilayer Coatings for Tribology Applications

MoS_2 coatings exhibit excellent lubrication performance under high dry or vacuum conditions due to the easy shear between lattice layers [56][57]. However, when rubbed in humid air, the dangling bonds at the edge of MoS_2 react strongly with O, resulting in higher COF and shorter service life [58][59][60]. Aiming to the shortcomings, MoS_2-based multilayer coatings have been designed to further enhance the performance. The tribological behaviors of multilayer coatings of MoS_2 and metallic including Au, Ni, Pb or PbO were studied in humid air with 50% relative humidity, which exhibited lower and more stable COF compared to pure MoS_2 coating [61]. The function mechanism of metal for the sputter-deposited metal–MoS_2 multilayer coatings is believed to be the optimization of the MoS_2 structure. Kong et al. [62] investigated the tribological behaviors of MoS_2/Ti–MoS_2/Si multilayer coatings deposited by magnetron sputtering, indicating that better lubrication performance can be achieved by the multilayer design of coatings. Those results indicated that the multilayer structures have potential to improve the tribological behaviors of conventional MoS_2 coating, but the inherent mechanisms are still worth further investigation to guide the designing of MoS_2-based coatings for future application.

With the development of coating fabrication and characterization techniques, several new findings shed the light on the precision structure design of multilayer coatings from an atomic view [63][64]. Dwivedi et al. [65] developed C/SiN_x multilayer coatings with layer thickness of 7–8 nm using an enhanced atomic intermixing (formation of nanocomposite interfaces) approach, leading to 2–10 times better macroscale wear durability compared to conventional coatings with larger thickness of 20–100 nm. The enhanced performance can be attributed to the high sp^3 bonding of the carbon overcoat and increased interfacial strength induced by intermixing, leading to improved adhesion and robustness of the coatings. Khadem et al. [66] designed discreate periodic nanolayered coatings, which had a different structure compared to conventional multilayer coatings. The discrete periodic nanolayered coatings exhibited better wear-reduction performance compared to conventional multilayer coatings, which can be attributed to the reduced interfacial defects. The tribological performance was further improved by surface-texturing treatment. Advanced research tools make it possible to investigate the fundamental mechanisms of multilayer coatings in tribological application.

Two-dimensional (2D) materials, including graphene-family materials [67][68][69][70][71][72], MoS_2 [58][59], and black phosphorus [73][74][75][76] have been used as lubricants because of their low interlayer shear strength. Recently, multilayer coatings with 2D materials have also been designed to promote tribological properties. Most recently, Fan et al. [77][78][79][80][81][82] fabricated coatings with $Ti_3C_2T_x$ Mxene and achieved excellent self-healing, antiwear, and anticorrosion capacity. Saravanan et al. [83] fabricated multilayer coatings with graphene oxide and PEI via layer-by-layer assembly technique. Macroscale superlubricity (COF < 0.01) can be achieved with the multilayer coating having a thickness of about 300 nm. The superlubricity mechanism is believed to be the formation of carbon nanoparticles in dry conditions. In the subsequent study, it was found that the formation of transfer layer is also critical for the achieving of ultralow friction [84]. Achieving macroscale superlubricity is possible with multilayer coatings containing 2D materials, but the environment adaptivity still needs to be improved, and the inherent mechanisms also need to be further investigated.

5. Mechanisms for Controlling Friction and Wear Using Multilayer Coatings

Coatings have been widely used in industrial applications as protection for cutting tools, dies, pistons, etc. However, the performance of monolayer coatings is usually restricted by their poor adhesion with substrate, and the high residual stress induced by the fabrication process. In addition, the mismatch of the mechanical properties between substrates and coatings also suppresses the performance of monolayer coatings. Aiming to solve these problems, the concept of multilayer coating has been proposed; and lots of work has been carried out to enhance the coating performance through the multilayer structure. One of the fundamental concepts of the multilayer design is stress relaxing and crack deflection [39]. Back in the 1970s–80s, researchers attempted to build multilayer coatings through alternating thin layers with high-shear-modulus and thin layers with low shear modulus based on the models of Hoehler. In 1990, Holleck et al. [39] found that the multilayer structure of TiC/TiB_2 coatings leads to the deflection of cracks through the interface zones, causing energy dissipation without coating failure. Layer thickness also has influence on the stress distribution of the multilayer coatings. In addition, from an engineering point of view, when a normal force is applied on the coating's surface, the multilayer coating with thin, soft layers can reduce the maximum bending stress. With the multilayer design with soft and hard layers, the plastic yielding of hard layers can be avoided, especially under the condition with cyclic loading and fatigue. Another fundamental concept for the designing of multilayer coating is the functional design of different layers for purposes such as adhesion, load supporting, lubrication, and wear reduction, etc. [45]. The wear-resistance and lubrication performances can be enhanced through the multilayer design of the coatings. However, macroscale friction is a complex physical–chemical process. The friction and wear-reduction mechanisms of multilayer coatings with different structures and compositions are different. Hence, various mechanisms for controlling friction and wear using multilayer coatings have been proposed (**Table 1**), which can guide the future development of the multilayer coating systems.

Table 1. Friction and wear-reduction mechanisms of multilayer coatings.

Types of Multilayer Coatings	Preparing Methods	Lubrication Properties	Friction-Reduction Mechanisms	Wear-Reduction Mechanisms
TiN/Ti [85]	Large area filtered cathodic arc deposition	COF reduced from 0.82 (TiN) to 0.6 (with Ti layers thickness of 25 nm)	Lower shear strength of soft Ti layers	—
Ti/TiN [86]	High-vacuum magnetron sputtering	COF reduced from 0.54 (TiN) to 0.48	Formation of $TiAlN_xO_y$ and TiN_xO_y tribolayers	—
TiN/CrN [87]	Reactive magnetron cathodic sputtering	COF reduced from 0.9 (TiN) and 0.6–0.7 (CrN) to 0.3–0.5	Enhanced hardness and formation of the dense Cr_2O_3, and CrO_3 oxide layer	
CrN_{HIPIMS}/TiN_{DCMS} [88] *	High-power impulse magnetron sputtering (HIPIMS) and DC unbalanced magnetron sputtering (DCMS)	COF reduced to 0.05	Formation of humidity-triggered layers during dry-sliding tests under humid conditions	—
(Ti–Cr)N [89]	Cathodic arc deposition	COF reduced from 0.7 (TiN) and 0.75 (CrN) to 0.4	Formation of mixed-phase films with plastic deformed wear debris	

Types of Multilayer Coatings	Preparing Methods	Lubrication Properties	Friction-Reduction Mechanisms	Wear-Reduction Mechanisms
CrTiN/TiCN and CrTiN/CrCN [90]	Cathodic arc PVD	COF reduced from 0.8–1.0 (bare substrates) to 0.2	Graphitization of the amorphous carbon phase	Improved adhesion between individual layers; increased coating hardness; graphitization
CrN/DLC/Cr-DLC [50]	PECVD	COF reduced to 0.087	Lubrication of DLC; supporting of CrN layers; enhancement of crack-propagation inhibition; increased elastic recovery capability	
Multilayer DLC with hard and soft layers [55]	Unbalanced closed-field magnetron sputtering	Lower COF during running-in process with soft top layer	Formation of transfer layer with soft top layer to provide low friction and wear	
MoS_2/Ti–MoS_2/Si [62]	Unbalanced magnetron sputtering	COF reduced to 0.0432	Improved compactness and orientation of MoS_2; improved oxidation and moisture resistance of MoS_2; higher hardness; hindered dislocations motion and crack propagation	
C/SiN_x overcoats [65]	Magnetron sputtering in situ with carbon deposition; high-energy carbon treatment	COF reduced from 0.4 (bare substrates) to lower than 0.2	Extremely high adhesion governed by atomic intermixing, sufficient carbon thickness; high sp^3 bonding	
Polyethylenimine/graphene oxide [83] *	Layer-by-layer deposition	COF reduced from 0.60	Reduction in the contact area due	—

References

Types of Multilayer Coatings	Preparing Methods	Lubrication Properties	Friction-Reduction Mechanisms	Wear-Reduction Mechanisms
		(substrate) to lower than 0.01	to the formation of carbon nanoparticles in dry conditions	

* The influence of environment humidity on the tribological behaviors was investigated

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