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Review

Solid–Liquid Composite Lubrication (SLCL) Based on Diamond-Like Carbon (DLC) Coatings and Lubricating Oils: Properties and Challenges

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Abstract: In the field of industrial lubrication, solid–liquid composite lubrication (SLCL) techniques based on diamond-like carbon (DLC) coatings and lubricating oils are emerging recently, which may be applied in many fields in the near future, especially automotive industries. The tribological behaviors of SLCL systems depend strongly on the compatibility between DLC coatings and oils. This review describes the advantages of SLCL techniques by pointing out the synergistic effects between DLC coatings and lubricating oils. Then the main factors determining the tribological performance of SLCL systems are discussed in detail. Finally, a conclusion about the characteristics of reported SLCL systems is made, and a prospect about the potential development of SLCL technology is proposed. On the basis of the relevant literature, it could be found that the tribological properties of SLCL systems were influenced by many more factors compared with individual DLC lubrication or individual oil lubrication due to the complicated tribo-chemical reactions involving DLC and oil during friction. And under some optimized working conditions, the tribological performances of SLCL systems (friction and wear reduction) are superior to individual DLC lubrication and individual oil lubrication. However, the tribological performance of SLCL systems needs to be further improved (for example, to achieve superlubricity and ultra-low wear simultaneously) by adjusting the structures of DLC coatings, regulating the compositions of oils, and most importantly, enhancing the physicochemical and tribological synergies between DLC coatings and oils. This review provides a comprehensive understanding of the SLCL technology, which may be very helpful for the researchers and engineers in the field of industrial lubrication and tribology.

Keywords: solid–liquid composite lubrication; DLC; oils; friction; wear

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1. Introduction

One sixth to one third of the fuel is consumed by friction in passenger cars [1], and about one third of the fuel is used to overcome friction in heavy-duty vehicles such as trucks and buses [2]. The reduction in friction in vehicles can save fuel and decrease CO_2 emissions [3]. Meanwhile, decreased wear can improve the durability of frictional components in vehicles. Consequently, scientists and engineers around the world are searching for advanced friction-reduction and anti-wear technologies, especially applicable in the widely used steel-based tribo-couples. The traditional method to reduce the friction and wear of steel vs. steel tribo-pairs is the utilization of lubricating oils (low-cost but efficient) at the interfaces [4,5], which has been accepted for many years in daily lives and industries. Researchers are always improving the lubrication performance of oils by adjusting their compositions and optimizing the compatibility between oils and tribo-pairs [6]. In the past decades, diamond-like carbon (DLC) coatings as surface modification materials attracted much attention due to their low friction [7], high wear resistance [8], and great potential as bio-compatible coatings used in the human body [9,10]. When sliding against steel, DLC coating exhibited an ultra-low coefficient-of-friction (COF) at

0.006 in vacuum due to the formation of multilayered spherical nanocluster structures [11]. This indicates the DLC coating serves as a self-lubricating protective layer in space-like conditions, where traditional oils cannot survive. Another advantage of the DLC coating is that it can maintain a very low COF at relatively high temperatures for a long time. For instance, Zeng et al. [12] observed that the ultra-low COF (~0.008) between DLC-coated steel ball and disc at a testing temperature up to ~600 °C could be maintained for about 200 min. The long-term and ultra-low COF was attributed to the synergism between the shielding effect of hydrogen in the DLC films and the formation of some oxides on the contact surface. An extremely low COF of 0.0016 was obtained for DLC coatings sliding against Al_2O_3 balls in dry air [13]. Tsigkis et al. showed the DLC coatings had a great potential to be applied to mechanical components in future missions to Venus due to the superb tribological properties under high pressure and high temperature CO_2 harsh conditions [14].

However, the tribological properties of DLC coatings are very sensitive to the change of environmental conditions, particularly the relative humidity (RH) [15,16]. Gong et al. revealed the COF of DLC coatings sliding against different counterparts (Al_2O_3, Si_4N_3, or steel) all increased with the increase in RH, and the COF stability was worse under higher RH [13]. In dry air, shear stress-induced multilayer graphene was responsible for the superlubricity of DLC coatings; however, under a high RH, the formation of graphene was limited because the dangling bonds of the graphene-like domains in the DLC coating were saturated by water molecules. Different from the COF trend, the wear of DLC coatings decreased with the increase in RH [17]. This was attributed to the formation of low energy surfaces by the water adsorption at the dangling carbon bonds on the edge-faces of the sp^2 clusters in the DLC coating.

Based on the above reports, it can be concluded that DLC coatings have some advantages compared with traditional oils when applied in some harsh conditions such as vacuum and/or high temperature; unfortunately, the tribological properties of DLC coatings depend strongly on RH, which is undesirable in practical applications. In practical engineering applications, low COF, wear resistance, and stable tribological performance under dynamic conditions are all desired. Though scientists around the world are trying to modify the DLC coatings for minimizing the influence of water molecules, no obvious improvements have been obtained yet. The dangling bonds in DLC tend to interact with water molecules easily and irreversibly due to the formation of C-O bonds, which leads to a significant deterioration in the tribological properties of DLC coatings, such as increased COF and enhanced wear.

On the other hand, the widely used lubricating oils are not sensitive to outer RH. And the water content in oils is often kept at a very low level when the oil functions due to the increase in oil temperatures. This means the oils can behave as a barrier that protects DLC coatings from moisture, keeping the tribological properties of the coating relatively stable. The combination or even synergy of solid DLC coatings and liquid lubricating oils, i.e., solid–liquid composite lubrication (SLCL), is desired to show improved environmental adaptability and tribological property based on the synergistic effects between the two components. Previous reports revealed the tribological performance of SLCL systems was often determined by the compatibility between DLC coating and oil additives [18]. Consequently, many scientists are trying to optimize the SLCL systems by adjusting the micro-structures of DLC coatings and/or tailoring the compositions of oils. Optimized SLCL systems with satisfactory tribological properties will be potentially applied in many fields, such as vehicles, ships, turbines, and manufacturing equipment.

In order to provide a comprehensive understanding about the history and future trends of SLCL systems, this paper gives a detailed review of the common SLCL systems, including both DLC coatings and lubricating oils, discusses their tribological properties, summarizes the lubrication and invalidation mechanisms, points out the bottlenecks that constrain the practical applications, and suggests some potential solutions to the bottlenecks.

2. Advantages of SLCL Systems Including DLC Coatings and Oils

2.1. Properties and Bottlenecks of DLC Coatings

DLC is a carbon-dominated meta-stable material composed of sp^3- and sp^2-hybridized bonds [19]. Depending on the hydrogen contents, DLC coatings can be categorized into two groups: non-hydrogenated and hydrogenated [20]. The mechanical and tribological properties of DLC coatings depend on the hydrogen content and the ratio of sp^3-hybridized bonds to sp^2-hybridized bonds (sp^3/sp^2) [21]. The sp^3/sp^2 ratio is determined by some parameters in depositing DLC (such as substrate bias voltage [22] and power applied [23]) and some post-treatment process [20]. DLC can also be classified into non-hydrogenated amorphous carbon (a-C), tetrahedral amorphous carbon (ta-C), hydrogenated amorphous carbon (a-C:H), and tetrahedral hydrogen-containing amorphous carbon (ta-C:H).

DLC coatings feature chemical inertness, high hardness, high plastic deformation, low friction, high wear resistance, and high corrosion resistance [24–27]. DLC coatings can be deposited on a wide range of materials and exhibit excellent compatibility with them, including steels [28,29], polymers [30,31], and alloys [32]. In automotive industries, the application of DLC coatings to gear could save the range of electric vehicles [33]. Compared with traditional lubricating oils having a typical COF in the range 0.1~0.15 when applied to metallic tribo-pairs, DLC coatings often exhibit a lower COF in the range 0.01~0.1 under dry friction conditions [34–36]. In particular, DLC coatings exhibit stable superlubricity (COF below 0.01) and long wear lives under some specific working conditions [37]. Superlubricity, a concept suggested thirty years ago [38,39], is a state featuring a COF lower than 0.01, an ultra-low level normal oils cannot reach. In some early literature, superlubricity phenomena were only limited to microscale or even nanoscale in some controlled environments [40], which seems far away from practical engineering applications. Very recently, macroscale superlubricity has been found in the sliding between steel pairs, in which the steel surfaces were usually modified by some lubricating coatings, such as a-C:H [41,42], under dry frictions. That means no liquids are used in these superlubricating systems.

The DLC lubricating coatings demonstrate exceptional thermal stability, making them effective for some high-temperature industrial applications where traditional liquid lubricants are ineffective. Si-doped DLC films can provide effective lubrication between Al_2O_3 balls and stainless steel at elevated temperatures (450 °C~500 °C) [43]. Wang et al. [44] revealed Si and N co-doped DLC coating had an ultra-low COF (0.033) at 500 °C because of the formation of compacted layers on the wear track. Vahidi et al. [45] found the hydrogenated DLC had a low COF around 0.05 when sliding against 100Cr6 steel balls at elevated temperatures (300 °C and 400 °C). In addition, the coating could protect the steel counterpart from severe wear even when the test temperature increased to 500 °C due to the consistent formation of tribolayers on the counterpart.

However, some previous reports show that the tribological properties of DLC coatings are very sensitive to atmosphere [46,47]. Zhang et al. [48] found DLC coatings sliding against SiC balls exhibited a much lower COF (0.06) and wear rate (2.2×10^{-9} mm^3/Nm) in nitrogen compared with that in oxygen (0.1 and 5.15×10^{-7} mm^3/Nm, respectively). Cui et al. [37] found the DLC film exhibited a lower COF in dry nitrogen compared with that in ambient air or in vacuum. Another disadvantage of the DLC coating is that, under either dry or lubricated conditions, its low friction often relies on surface graphitization [34,35]. This leads to rapid thinning of DLC coatings, resulting in a short lifetime [49].

2.2. Properties and Bottlenecks of Lubricating Oils

The COF between steel vs. steel tribo-pairs under the lubrication of traditional oils is often above 0.1 [50]. Some friction modifiers (such as organic compounds [51], inorganic nanoparticles [52], or inorganic–organic hybrids [53]) have been added into oils to reduce the COF. And recently, some lubricating oils (such as 1,3-diketone [54] and mixed lubricants [55,56]) have achieved superlubricity (COF < 0.01) between steel tribo-pairs. In these reports, the achievement of oil superlubricity often requires a long running-in period (>1 h) to realize surface conformity. In addition, the oil superlubricity depends strongly on specific operating conditions such as load and velocity [54]. That means the low-COF states may not be kept under some real dynamic conditions. Another bottleneck of traditional oils is the relatively low thermal stability due to the easily degradable organic molecules of the oils. Therefore, the oil lubrication needs to be improved, though it has been used in many fields, such as automotive applications.

2.3. The Necessity of SLCL

The combination of DLC coatings/lubricating oils and the fabrication of SLCL systems can utilize the advantages of DLC coatings (low COF) and oils (low wear and insensitivity to the outer atmosphere) simultaneously. The advantages of SLCL systems can be demonstrated in Table 1. Based on the advantages of SLCL systems compared with individual oil or DLC coating lubrication methods, the construction and measurement of various SLCL systems have been investigated intensively in recent years, as will be described in the following section.

Table 1. A comparison of the advantages and disadvantages of different lubrication methods.

Lubrication Methods	Advantages	Disadvantages
Oils	(1) Non-sensitive to outer atmosphere, (2) Low cost	(1) High COF, (2) Low thermal stability, (3) Not suitable for space applications
DLC coatings	(1) Low COF (even superlubricity), (2) High thermal stability, (3) Suitable for space applications	(1) High wear due to surface graphitization, (2) Sensitive to outer atmosphere
SLCL systems	(1) Non-sensitive to outer atmosphere, (2) Low COF and wear	(1) Not suitable for space applications, (2) Low thermal stability

3. Tribological Properties of SLCL Systems

This review focuses on the advances in SLCL techniques that combine DLC coatings and lubricating oils. Depending on the coating structures, oil compositions, counterpart materials, and working conditions, SLCL systems exhibit crucial variations in tribological behaviors. This part will outline the effects of some key parameters on the tribological properties of SLCL systems, including DLC coatings and lubricating oils.

3.1. Effects of Hydrogen Content in DLC

The Masuko group [57] compared the friction and wear characteristics of DLC coatings with different hydrogen contents (ta-C with zero hydrogen, a-C:10H with 10~15 at.% hydrogen, and a-C:30H with 30 at.% hydrogen) against bearing steel under the lubrication of additive-containing poly-alpha-olefin (PAO) oils. ta-C had a higher hardness (60~70 GPa) than the a-C:H coatings (~30 GPa). Wear acceleration of a-C:H in the presence of MoDTC additives in PAO was observed due to the formation of MoO_3 abrasive particles. However, MoDTC reduced the wear of both ta-C and uncoated steel. Traditional anti-wear agent ZDDP or a combination of ZDDP and MoDTC decreased the wear of all DLC coatings, especially the ta-C. In the presence of ZDDP+MoDTC, both MoO_3 and MoS_2 were detected. Vengudusamy et al. [58] investigated the influences of hydrogen contents in DLC coatings on friction and wear of DLC/DLC contacts lubricated by an API group III base oil. The a-C:H coating showed a higher COF than the a-C coating under the base oil, due to the higher sp^2 content in the a-C coating. Adding ZDDP to the base oil lowered the COF of the a-C:H coating, whereas an adverse trend was observed for the hydrogen-free a-C coating. Moreover, the addition of ZDDP to the base oil led to a decreased wear of the coatings.

3.2. Effects of Doping in DLC Coatings

A major problem the DLC coatings face when applied to various surfaces is their insufficient adhesion to substrates. This is mainly caused by the high internal stresses of DLC coatings and the mismatch of thermal expansion coefficients (TEC) between DLC coating and substrate [59]. Another problem that restricts DLC coatings considerably is the humidity sensitivity of the tribological behaviors. Numerous studies have reported that doping DLC with additional elements or phases may help to overcome these drawbacks. This section will outline simply some previous research about the effects of doping (with hetero-atom or nanocrystalline) in DLC coatings. Unfortunately, SLCL systems involving doped DLC coatings were rarely reported.

3.2.1. Hetero-Atom Doping

As reported, hetero-atom doping influences both mechanical and tribological properties of DLC coatings [60,61]. For instance, the research of Wei et al. [61] indicated Cr-doping or B-doping could decrease the residual stress significantly, while the doping had only a little influence on the hardness and elastic modulus of hydrogen-containing DLC coatings. Compared with un-doped DLC coatings, only a few studies reported the SLCL systems, including hetero-atom doped DLC coatings. This section will outline some typical reports about the construction and measurement of SLCL systems involving hetero-atom doped DLC films. In addition, some conclusions and discussions are also presented.

B Doping

Mori group [62] found, under a fully formulated gasoline engine oil containing Mo-type friction modifiers, the B-doped DLC coatings (B-DLC) had an extremely low COF below 0.03 in a coated steel block-on-steel ring tribo-pair. This low friction was attributed to the friction modifier-induced nano-scale smooth surface of the B-DLC coating and the formation of a selectively adsorbed friction modifier film. However, the Mo-type friction modifier increased the wear of the coating, indicating the Mo-type friction modifier was incompatible with B-DLC coatings. In order to reveal the interactions between B-DLC surfaces and Mo-type friction modifiers, Jusufi et al. [63] measured the adsorption isotherms of MoDTC and MoDDP (two widely used Mo-type friction modifiers in oils) on B-DLC surfaces with a quartz crystal microbalance. It was revealed that more negative free energy of adsorption (i.e., stronger adsorption) led to a reduced wear of the B-DLC surface. Due to its stronger adsorption, MoDDP showed a better anti-wear performance on the B-DLC surface compared with MoDTC. In addition, MoDDP-containing oil resulted in a low COF of around 0.04 for the B-DLC coating. The higher affinity of MoDDP to the B-DLC surface originates from its more polar regions (a polar region around Mo atoms and two polar regions around the phosphate groups) than MoDTC (only one polar region around Mo atoms). This result indicates the sulfur- and phosphorus-containing MoDDP is more compatible with the B-DLC coating than the sulfur-containing but phosphorus-free MoDTC, due to the excellent anti-wear performance of MoDDP.

N Doping

Nitrogen-doped DLC films (N-DLC) own very high hardness, close to diamond [64]. Liu et al. prepared nitrogen-doped ta-C coatings (ta-CNx) by introducing nitrogen gas to an ion beam-assisted electric beam deposition (IBA-EBD) system, and the tribological properties of the ta-CNx coatings were measured by sliding against steel under PAO base oils [65]. As shown in Figure 1a, the coating with a high nitrogen content (ta-CNx2) exhibited a lower wear than the un-doped ta-C and the ta-CNx with a low nitrogen content (ta-CNx1). Figure 1b indicates the ta-CNx2 coating shows better resistance to the increase in the oil temperature compared with the other two coatings. One reason is that the nitrogen doping could reduce the thermally activated tribo-chemical wear. Another reason is that the low surface roughness of the ta-CNx coating with a high nitrogen content makes mixed lubrication happen more easily, resulting in lower contact load and therefore better wear resistance [66].

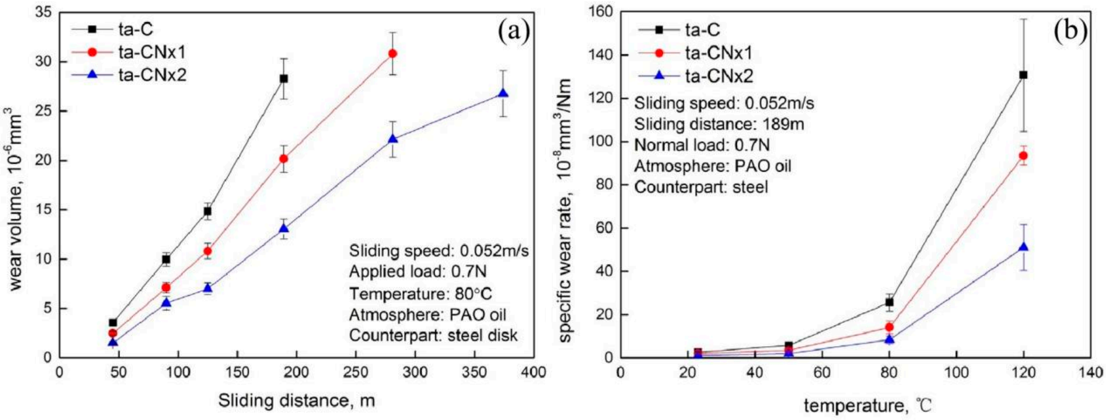


Figure 1. Wear properties of ta-C and ta-CNx coatings against steel under PAO base oil as a function of sliding distance (**a**) and as a function of oil temperature (**b**) [65].

F Doping

The changes in DLC coatings induced by F doping include reduced sp^3/sp^2 ratio, reduced Young's modulus and hardness, decreased residual stress, increased adhesion strength, lowered surface energy, enhanced corrosion resistance, and decreased COF [59,67]. The precursor for preparing F-doped DLC (F-DLC) is often CF_4 gas [67]. Compared with un-doped DLC, the friction of F-DLC with higher hydrophobicity showed a higher resistance to humidity [68]. The research by Wang et al. [69] revealed F-DLC coating exhibited an excellent tribological property with low COF and low wear in the stroke-physiological saline solution at 37 °C, showing potential applications of the coating in medical devices.

Si Doping

Si doping can alter the micro-structures and properties of DLC coatings remarkably. Zhang et al. [70] found Si doping of a-C:H coatings could: (1) increase the sp^3/sp^2 ratio by preferentially substituting the sp^2-hybridized carbon atoms, (2) improve the hardness and adhesion strength while reducing the residual stress of the coating, (3) enhance the thermal and mechanical stability through inhibiting the graphitization at elevated temperatures, and (4) improve the high-temperature tribological properties (up to 450 °C) in air due to the formation of Si-containing tribo-layers. Yu et al. [71] also observed the low-friction phenomenon of Si-doped DLC coatings against Al_2O_3 counterparts under high temperatures up to 500 °C. The formation of Si-containing species was responsible for the low friction at high temperatures. Kassim et al. [72] compared the tribological behaviors of a-C:H and Si-doped a-C:H coatings against steel under PAO with MoDTC additives. The average COF values of un-doped and Si-doped a-C:H were around 0.05, a level lower than most lubricating oils. The specific wear rate of the Si-DLC coating was only half of the un-doped DLC under PAO or MoDTC-contained PAO. One of the reasons may be the higher hardness of Si-DLC (25 GPa) than the un-doped DLC (18 GPa).

Ti Doping

Guo et al. [35] prepared Ti-doped DLC coatings (Ti-DLC) by a hybrid ion beam deposition system. Under boundary lubrication, the Ti-DLC coating with a high Ti concentration (27 at.%) showed a very low wear rate. In contrast, this coating was worn out quickly under dry friction due to abrasive wear.

Cu Doping

Cu atoms have a low solubility in carbon matrix, so the maximum doping concentration of Cu in DLC films cannot exceed 2 at.% [73]. Binjua group [74] investigated the friction and wear of ta-C coatings and copper-doped ta-C coatings (Cu/ta-C) under the lubrication of PAO with or without MoDTC additives. Tribological experiments at room temperature revealed that, compared with the Cu/ta-C coatings, the ta-C coatings against Si_3N_4 balls exhibited a lower COF and higher wear resistance under the MoDTC-containing PAO oil. Under the lubrication of PAO without MoDTC additives, the average COF values of ta-C and Cu/ta-C were similar; however, the Cu/ta-C coating showed a much lower wear compared with ta-C. It seemed that the Cu doping was not helpful to reduce the friction of ta-C coatings but decreased the wear of coatings under PAO and the wear of counter balls under both types of oils.

W Doping

Some papers reported the enhanced high-temperature (up to 500 °C) tribological properties of DLC coatings under dry conditions by W doping [75,76]. This is mainly due to the formation of a tungsten oxide layer at the interface. Under lubricated conditions, the working temperature cannot reach such high levels because the oil would decompose quickly. The Austin group [77] compared the wear of a-C:H and W-doped DLC (W-DLC) against a cast iron counterpart under the lubrication of a fully formulated oil containing ZDDP and some other additives (detergent, dispersant, and antioxidant). a-C:H coating exhibited a lower wear rate than W-DLC. A glassy phosphate film was formed between the a-C:H coating and cast iron. The failure of W-DLC was attributed to the generation of η-phase $W_{6\text{-}x}Fe_xC$. Vengudusamy et al. [58] investigated the effects of W-doping on the mechanical and tribological properties of a-C:H coatings in DLC/DLC tribo-pairs under an API group III base oil. The a-C:H:W coatings showed a lower hardness and elastic modulus compared with the a-C:H coatings. As shown in Figure 2, under the base oil, the a-C:H:W coatings experienced a much higher wear than both the a-C:H coating and uncoated steel. Addition of ZDDP to the base oil can lower the wear of both a-C:H and a-C:H:W coatings due to the formation of ZDDP-derived tribo-films containing Zn, S, and P elements. This study indicates there are no advantages in both mechanical and tribological properties for the W-doped a-C:H coatings compared with un-doped a-C:H coatings.

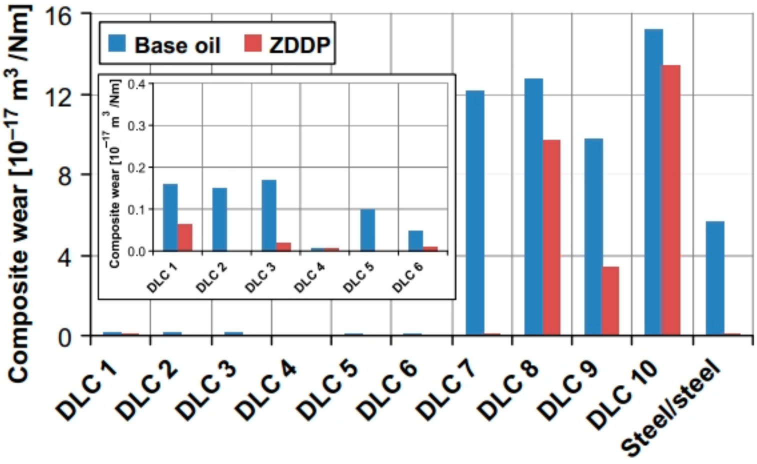


Figure 2. Comparison of composite wear coefficients for different types of DLC/DLC tribo-pairs tested in base oil (an API group III oil) and ZDDP solution. DLC 1 is a-C, DLC 2~6 mean a-C:H coatings with increasing hydrogen concentrations (17~25 at.%), and DLC 7~10 mean a-C:H:W coatings with a fixed hydrogen concentration (15 at.%) and increasing W concentrations (12~21 at.%) [58].

Co-Doping with Different Hetero-Atoms

Co-doping with two or even more kinds of hetero-atoms is an effective method to combine the desired properties of different elements to modulate the micro-structures and improve the tribological behaviors of DLC coatings [44,78]. For instance, Yu et al. [79] found Si and W co-doped DLC coatings showed very well high-temperature (up to 500 °C) tribological properties when sliding against Al_2O_3 under dry conditions, due to the synergistic effects of film graphitization, improved mechanical properties, and the formation of W-containing and Si-containing species on the wear track. The co-doping of F and Si deteriorated the mechanical properties but reduced the COF greatly of hydrogenated DLC (H-DLC) coatings [80]. Compared with un-doped a-C and Si-doped a-C, Si and Al co-doped a-C coatings presented lower tribological moisture sensitivity under dry friction against steel [81]. Under low RH, compacted graphitized tribo-films were formed; under high RH, colloidal silica tribo-films with low shear strength were produced. The introduction of Al can relax internal stress and improve the adhesive strength of the a-C coating.

Based on the above descriptions, it may be concluded that the tribological properties under dry friction of some doped DLC coatings are superior to those of un-doped coatings due to the physicochemical changes induced by the hetero-atoms incorporation. However, research about SLCL systems that contain hetero-atom-doped DLC coatings is very lacking so far.

3.2.2. Nanocrystalline Doping

Ren et al. [82] investigated the effects of WC nanocrystallines in a-C coatings on the tribological properties of an SLCL system, including a tribo-pair of steel/a-C coating and a PAO oil with or without ZDDP. The introduction of WC resulted in no obvious changes in COF under the lubrication of PAO or PAO+ZDDP. Under PAO, the wear rate of a-C coating was increased by the WC doping due to the lower hardness of the WC-doped a-C coating (14.3 GPa) compared with the un-doped coating (17 GPa) and intensified abrasion of the coating by hard WC particles. The addition of ZDDP into the PAO oil lowered the wear of both a-C and WC-doped a-C coatings due to the formation of ZDDP-derived phosphorus- and sulfur-containing tribo-films, which acted as a protective barrier between the tribo-contacts. Under PAO+ZDDP, the wear rate of the WC-doped coating was lower than the un-doped coating. This was attributed to the generation of anti-wear WS_2 and WO_2 by tribo-chemical reactions.

3.3. Effects of DLC Functionalization

The reported well-established methods used to functionalize the surfaces of DLC coatings include laser texturing [83], atomic oxygen (AO) erosion [84], and ultra-violet (UV) irradiation [85]. AO and UV are typical space environment factors, both making the mechanical properties of DLC coatings decrease.

AO erosion was found to increase the residual stress while decreasing the hardness and elastic modulus of hydrogen-containing DLC films due to the erosion-induced reduction in the sp^3 hybrid bond content [61]. UV irradiation can decrease the hardness and elastic modulus while increasing the residual stress and surface roughness of a-C:H coatings, due to breakage of chemical bonds in the coatings [86]. And UV decreased significantly the wear life of the coating sliding against steel under vacuum conditions. The effects of UV irradiation to a-C:H on the tribological properties of a tribo-pair SUJ2 ball vs. a-C:H coating under the lubrication of PAO oil containing traditional additives MoDTC and ZDDP were investigated by the Taib group [85]. The UV irradiation created a graphite-like layer on the topmost surface of the a-C:H coating. During frictions, the functionalized surface attracted more additives and produced a thicker tribo-film between the tribo-pairs compared with the as-deposited coating. Consequently, the UV-irradiated a-C:H coating showed a lower COF than the as-deposited coating.

Surface texturing of tribo-pairs is an effective technique to improve the tribological properties [87,88]. Surface texturing of DLC coating on a polymer matrix was fabricated by Marian et al. [89]. Firstly, PMMA with single- and multi-scale surface textures was prepared by 3D printing. Then, an a-C:H coating was deposited on the polymer surface. Consequently, the obtained coating had the same textures as the 3D-printed polymer matrix. Tribological tests indicated that, under dry sliding conditions, the 3D printed textures and the DLC coatings had synergistic effects, leading to reduced friction and wear of the polymer surface.

Another DLC functionalization route is functionalizing the interface between DLC coating and the substrate. For example, Brittain et al. [90] used graphene nanoplatelets (GNP, multi-layer graphene materials) to functionalize the interface between DLC coating and substrate (a high-speed steel (HSS) coated with a Cr/WC/W layer). The schematic flow diagram for the synthesis is shown in Figure 3. The interface functionalization with GNP reduced the COF from above 0.07 to only 0.03 when the DLC film slid against a cast iron counter-body under a PAO base oil lubrication. The significant reduction in both friction and wear was attributed to the highly graphitic transfer film with a low shear strength on the counter-body. Using a similar pathway, Wei et al. [91] used multi-wall carbon nanotubes (CNTs) to functionalize the DLC/silicon interface. The introduction of CNTs increased the hardness and elastic modulus of DLC coating while reducing its residual stress and COF.

3.4. Effects of DLC Surface Morphology

In an SLCL system involving a DLC coating and an engine oil containing both MoDTC and ZDDP, a rough DLC surface may promote MoS_2 formation. Komori et al. [92] deposited DLC on micro-shot peened rough surface substrates and got “pre-structured” DLC coatings. Compared with the DLC deposited on polished substrates, the “pre-structured” DLC coatings showed lower COF and lower wear. The surface structuring also inhibited the DLC wear by reducing the Mo-oxides and Mo-carbides production and limiting the coating transformation to graphitic structures.

3.5. Effects of Oil-Soluble Additives in Oils

Commercial engine oils usually include base oils and some oil-soluble additives to adjust the physicochemical properties of the oils. These additives often include antioxidants, anti-wear agents, friction modifiers, detergents, dispersants, antifoaming agents, antirust agents, antiseptics, and viscosity index improvers. Therefore, the effects of oil-soluble additives on the tribological properties of SLCL systems need to be investigated deeply before an SLCL system is commercialized. Though a few reports show the effect of additives is minor [93], most relevant papers have found the significant influence of oil additives on the tribological properties of SLCL systems. The most widely used anti-wear agent is ZDDP, which can form a dense tribo-film separating the tribo-contacts effectively [94]. Some oil-soluble organic molybdenum additives have been explored intensively as friction modifiers in lubricating oils in recent years because they can form a MoS_2-containing tribo-film during friction [95,96]. As the tribological properties of oils are mainly determined by friction modifiers and anti-wear agents, this section will outline the effects of them on the tribological properties of SLCL systems.

3.5.1. ZDDP

ZDDP is the most widely used anti-wear and extreme-pressure agent in lubricating oils applied to steel vs. steel tribo-pairs [97,98]. And it is often used in the oils of SLCL systems [99]. The anti-wear performance of ZDDP results from the formed protective films on rubbing surfaces [100].

In an SLCL system consisting of a steel/a-C coating tribo-pair and PAO base oil [82], the introduction of ZDDP to the oil made the COF more stable because the ZDDP-derived zinc polyphosphate layer could digest some abrasive particles from the rubbed surface [101].

In addition, the a-C coating showed a lower wear under PAO+ZDDP compared with that under PAO. This was due to the protective barrier effects of the ZDDP-derived tribo-films that can suppress tribo-induced surface graphitization of the a-C coating.

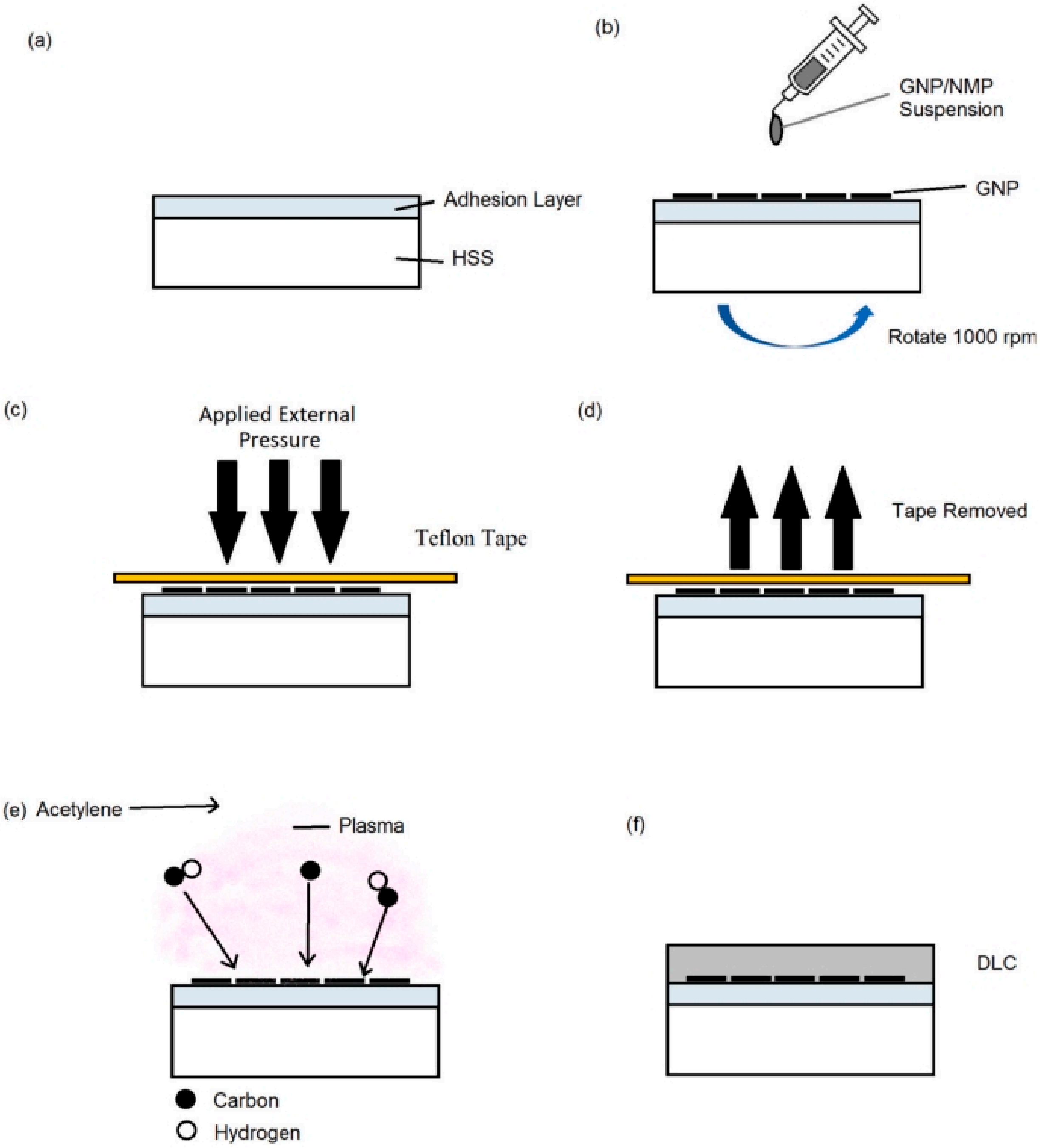


Figure 3. Schematic flow diagram for the DLC/substrate interface functionalization: (**a**) HSS substrate with an adhesion layer Cr/WC/W, (**b**) drop cast spin coating of GNP suspension on the surface (GNP was dispersed in N-methyl-2-pyrrolidone, NMP), (**c**) Teflon tape pressed on the GNP-coated surface and then heated at 200 °C for 3 h to improve the adhesion of GNP to the substrate, (**d**) tape removal, (**e**) DLC deposition by PECVD, and (**f**) the structure of the product in which the DLC/substrate interface was functionalized by GNP discrete islands [90].

A problem in SLCL systems consisting of a hydrogenated DLC coating and a Mo-containing oil is the Mo-accelerated wear of the coating [102,103]. Mechanical wear, chemical wear, and graphitization-induced delamination were often used to explain the wear mechanisms [104,105]. DLC coatings are often microns thick, so a small amount of wear may erase away the thin DLC films quickly, limiting the coating lifetimes. An effective solution is to add some ZDDP additives into the Mo-containing oil, which can form a protective layer during friction, limiting the wear of DLC coatings. For example, Chen group [106] found the friction and wear of DLC coatings could be reduced significantly when using a di-isooctyl sebacate (DIOS) base stock incorporated with ZDDP and molybdenum polyisobutylene succinimide (MPIBS)-modified molybdenum oxide nanoparticles (MONPs) to lubricate a steel/DLC coating pair. The MPIBS agent not only behaved as a dispersant to disperse the MONPs in the base oil but also produced MoS_2 through tribo-chemical reactions with ZDDP. As a result, a composite tribo-film consisting of MONPs, MoS_2, and ZDDP-derived amorphous polyphosphate was formed, which reduced both the friction and wear effectively. The DIOS oil without additives produced a COF at ~0.10 and a wear rate of 0.4×10^{-9} $mm^3/(Nm)$, while the addition of 0.7 wt.% MPIBS-MONPs and 0.8 wt.% ZDDP led to a COF at only ~0.03 and a wear rate of only 0.05×10^{-9} $mm^3/(Nm)$.

3.5.2. MoDTC

The mainstream friction modifier in commercial engine oils is phosphorus-free and sulfur-containing molybdenum dithiocarbamate (MoDTC), a type of oil-soluble organic molybdenum product [107,108]. The superb friction-reduction function of MoDTC originates from the formation of MoS_2-containing tribo-films [109,110]. As a widely used friction modifier in commercial oils, the influences of MoDTC on the tribological properties of SLCL systems have been measured for years [111]. However, numerous reports have shown that MoDTC in oils was antagonistic to some DLC coatings, especially the a-C:H coatings in steel/a-C:H contacts [112]. This section will introduce some typical reports about SLCL systems containing DLC coatings and MoDTC additives in oils, the basic mechanisms of wear acceleration that were often observed in MoDTC-lubricated DLC coatings, and some potential solutions to enhance the compatibility between MoDTC additives and DLC coatings in SLCL systems.

Friction Reduction but Wear Acceleration Induced by MoDTC

MoDTC has been one of the essential additives in lubricating oils. Therefore, understanding the compatibility between DLC coatings and MoDTC in oils is very important for the industrial applications of SLCL techniques. Numerous previous studies have observed that MoDTC often imposed a high wear on DLC, especially hydrogenated DLC coatings. On the other hand, a reduced friction of the DLC under MoDTC-containing oil was frequently recorded.

Vengudusamy et al. [111] compared the tribological properties of seven types of DLC coatings (a-C, a-C:H, a-C:H:W, a-C:H:WC, Si-DLC, ta-C, and ta-C:H) in two types of contacts (DLC-coated ball/DLC-coated disc and steel ball/DLC-coated disc, respectively) lubricated by an API Group III oil containing 0.3 wt.% MoDTC. It was found that MoDTC reduced the COF of both DLC/DLC and steel/DLC contacts; MoDTC did not affect the wear resistance of DLC/DLC contacts; however, MoDTC promoted the DLC wear in most steel/DLC contacts. As shown in Figure 4(left), under the lubrication of oils with or without MoDTC, the composite wear of a-C, a-C:H, Si-DLC, and ta-C:H self-mated tribo-pairs was extremely low, comparable to that of the steel/steel contact under the lubrication of MoDTC-containing oil. As shown in Figure 4(right), under the base oil, the composite wear of a-C/steel, a-C:H/steel, and Si-DLC/steel contacts was extremely low; however, MoDTC increased the wear significantly, especially for the a-C:H/steel contact. For the a-C:H:WC/steel and ta-C:H/steel contacts, MoDTC also increased the wear to some extent. For the ta-C/steel contact, the composite wear did not change due to MoDTC. For the a-C:H:W/steel contact, MoDTC lowered the wear to half of the value in the absence of MoDTC. Comparing these two figures, the composite wear of the DLC/steel contacts under MoDTC solutions was much higher than the corresponding DLC/DLC contacts except for the a-C:H:W and ta-C coatings.

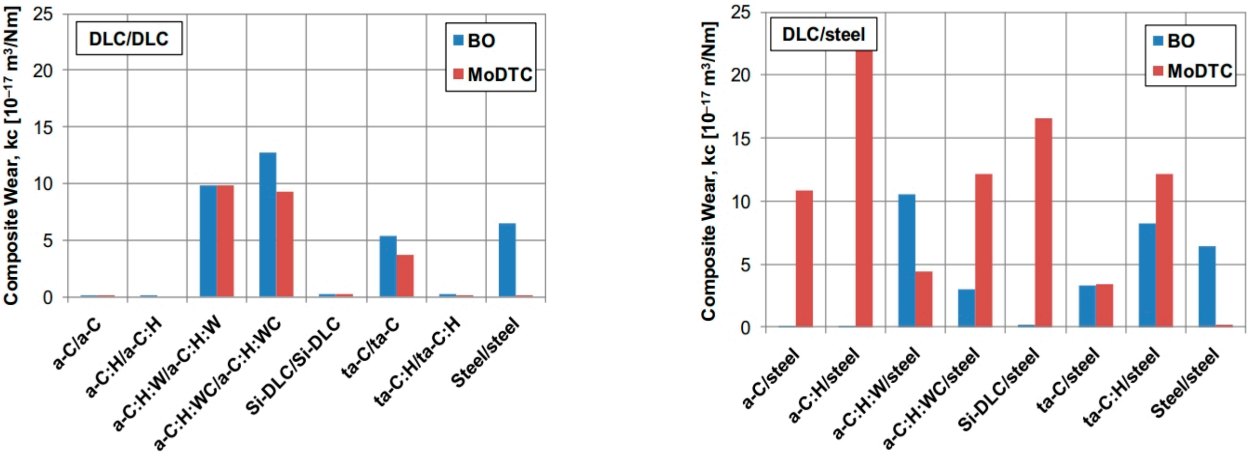


Figure 4. Composite wear of DLC/DLC tribo-pairs (**left**) and DLC/steel tribo-pairs (**right**) tested in base oil (BO) and MoDTC solutions [111].

Okubo et al. [113] monitored the surface transformation of DLC coatings (a-C:H and ta-C) by in situ Raman tribometer. As shown in Figure 5a, under PAO+MoDTC, the intensity ratio between D peak and G peak (I_D/I_G) of the a-C:H coating decreased gradually from 1.5 to 0.5, indicating the formation of Mo_2C. On the other hand, the I_D/I_G of a-C:H coatings under PAO or PAO+MoDTC+ZDDP was stable throughout the tests, indicating the structural transformation of a-C:H was inhibited. As shown in Figure 5b, under PAO, the I_D/I_G of the ta-C coating increased gradually with sliding time, indicating the friction-induced graphitization of the ta-C surface [114]. For the ta-C/steel pairs under different oils, no differences between their friction and wear behaviors were observed. However, for the a-C:H/steel pair, wear acceleration by MoDTC and wear inhibition by ZDDP were observed. The additive package of MoDTC+ZDDP showed a little weaker friction-reduction performance compared with MoDTC for the a-C:H/steel pair. The hard Mo_2C-containing tribo-films contributed to the abrasive wear of a-C:H coatings under PAO+MoDTC.

Mechanisms of MoDTC-Accelerated Wear of DLC Coatings

As the SLCL systems, including DLC coatings and MoDTC additives in oils, are investigated intensively, the mechanisms of the DLC wear acceleration are gradually being revealed. It was indicated that MoDTC-derived compounds (i.e., Mo-carbides and Mo-oxides) contributed to the accelerated wear of DLC coatings. For instance, Mo-oxides with a sharp edge crystalline solid structure can result in mechanical wear of DLC coatings in steel/DLC contacts [115].

By an in situ Raman tribometer, Okubo et al. [116] found the wear acceleration of a-C:H films in the presence of MoDTC originated from both chemical wear and abrasive wear. The chemical wear was caused by chemical reactions between the a-C:H coating surface and some MoDTC-derived compounds, producing Mo-carbides particles; the abrasive wear was caused by the formed hard Mo-carbide particles. The mechanism of the wear acceleration is illustrated in Figure 6. At the a-C:H/steel contact, the MoDTC-derived compounds (MoO_3 and MoS_2) were reduced to Mo by the iron on the steel surface; then the Mo reacted with the dangling bonds of the carbon atoms on the DLC surface, producing Mo-carbides (Mo_2C) and promoting the chemical wear of DLC films; the hard Mo-carbide particles can also lead to abrasive wear of DLC films. In this pathway, the steel surface is essential for the wear acceleration of the a-C:H coating.

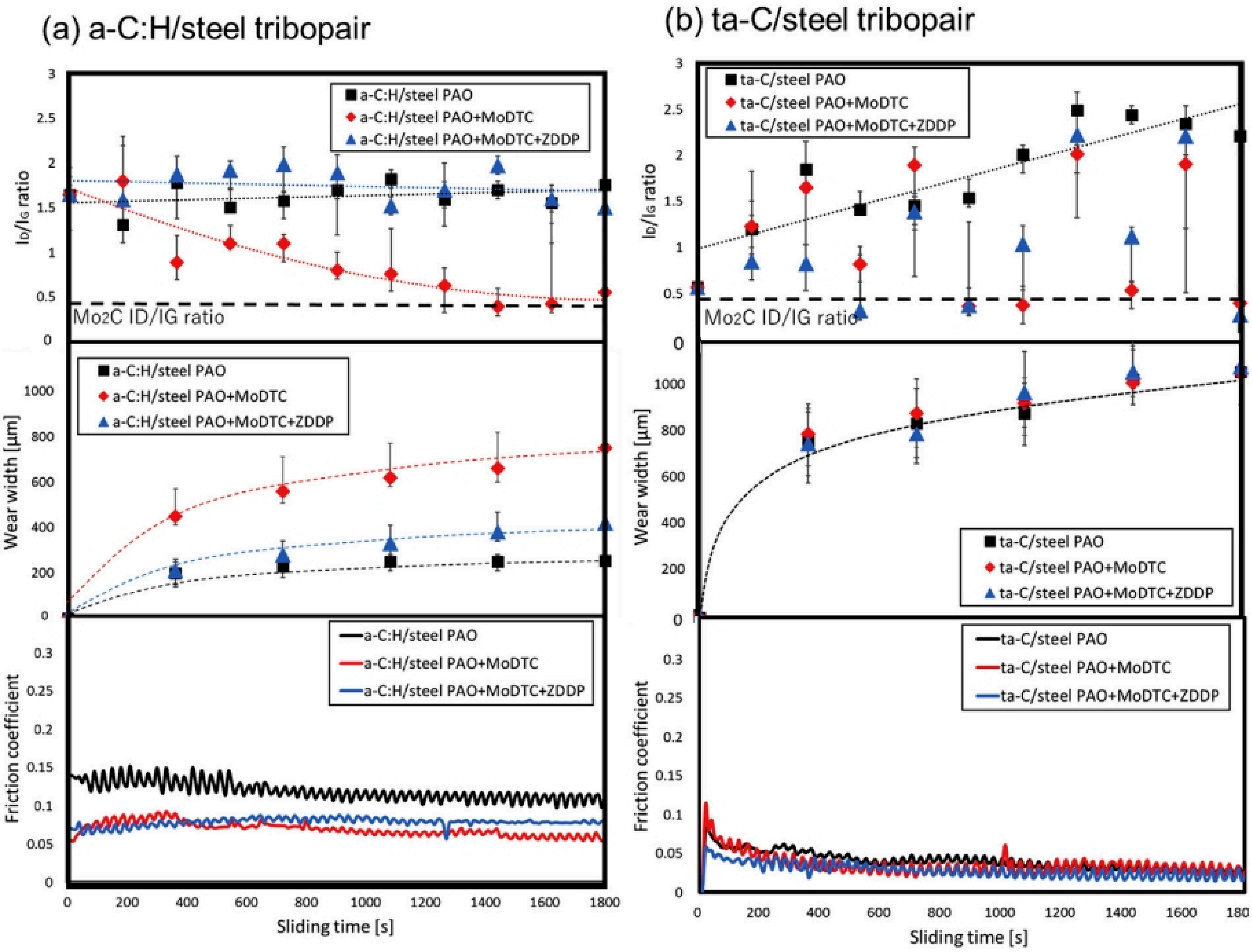


Figure 5. I_D/I_G ratios, wear widths of the coatings, and COF of the DLC coating/steel pairs lubricated by PAO, PAO+MoDTC, or PAO+MoDTC+ZDDP as a function of sliding time [113].

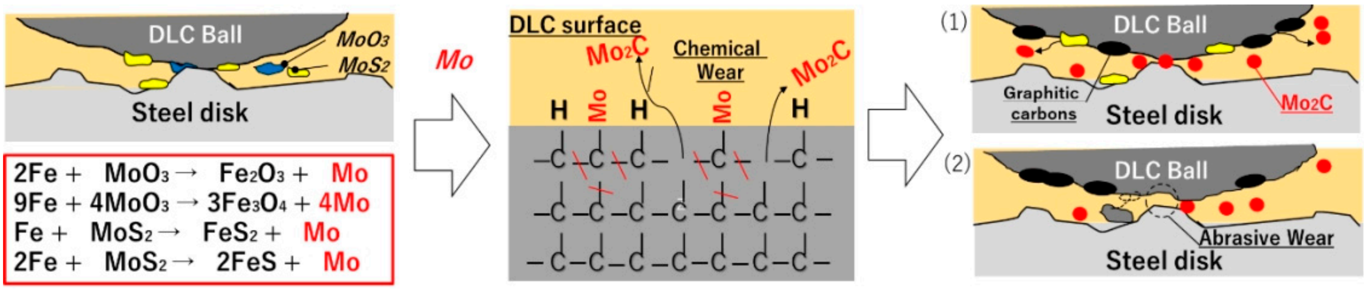
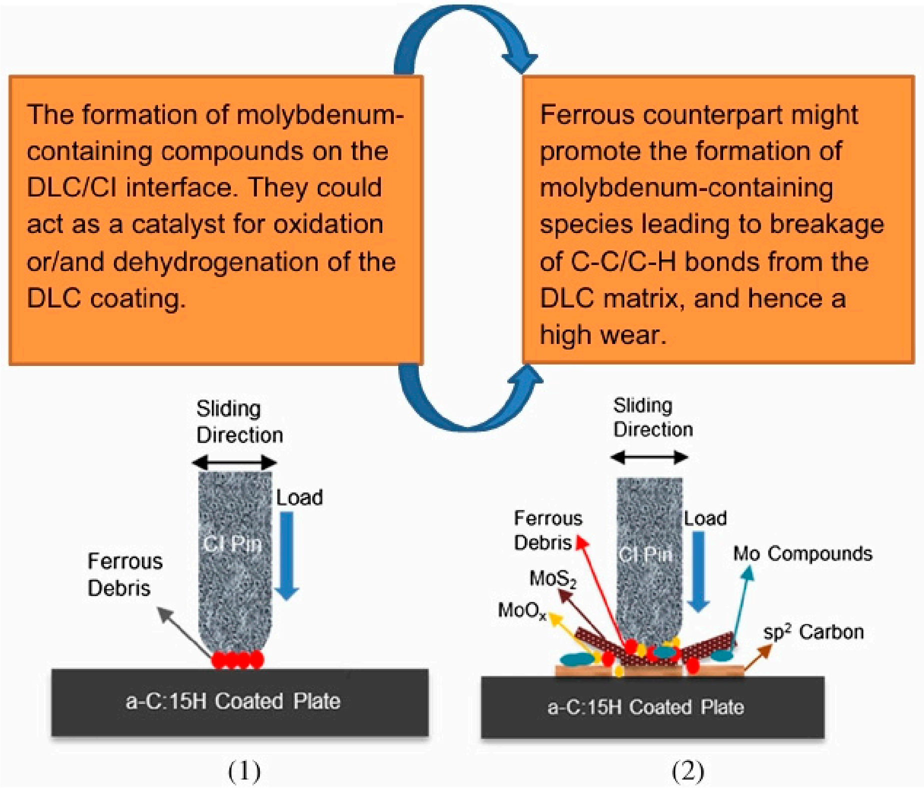


Figure 6. Schematic diagram of the mechanism of the wear acceleration of a-C:H films lubricated with MoDTC solution. In the right image, part (1) means the tribochemical reactions between Mo and DLC surfaces produce both graphitic carbons and Mo-carbides; part (2) means the hard Mo-carbides lead to the abrasive wear of DLC films [116].

Kosarieh et al. [117] proposed a wear mechanism of hydrogenated DLC induced by MoDTC additives. The schematic diagram of the wear mechanism is shown in Figure 7. Under tribological conditions, the moly-oxides formed from MoDTC could act as catalysts for oxidation and/or dehydrogenation of the DLC coating, leading to the wear of DLC. In addition, ferrous debris from the ferrous counterpart led to the formation of iron molybdates. The iron molybdates could accelerate DLC wear by breaking the C-C/C-H bonds.



The formation of molybdenum-containing compounds on the DLC/CI interface. They could act as a catalyst for oxidation or/and dehydrogenation of the DLC coating.

Ferrous counterpart might promote the formation of molybdenum-containing species leading to breakage of C-C/C-H bonds from the DLC matrix, and hence a high wear.

Figure 7. Schematic diagram of the MoDTC-induced wear mechanisms. (1) and (2) represent the first and the second stage of the wear mechanism. CI means cast iron, and a-C:15H represents 15 at.% hydrogenated DLC. The plate is made of high-speed steel [117].

Solutions to Inhibit the DLC Wear Induced by MoDTC

In an SLCL system, the accelerated wear of DLC coatings in the presence of MoDTC additives is of course harmful to the running machines. Some potential solutions have been proposed in the literature to lower the DLC wear while maintaining its low friction characteristics. They are listed as follows:

1. Using the additive package MoDTC+ZDDP instead of individual MoDTC

Haque et al. [118] found the wear acceleration effect of MoDTC on the DLC coatings in a cast iron/hydrogenated DLC tribo-pair can be canceled by the addition of traditional anti-wear agent ZDDP. The ZDDP-derived ZnO/ZnS compounds contributed to a lowered wear and enhanced durability of the hydrogenated DLC coatings. Ueda et al. [103] found the addition of ZDDP into a MoDTC-containing PAO oil inhibited significantly the coating wear in steel/a-C:H contacts, and the oil PAO+MoDTC+ZDDP led to a lower COF compared with PAO and PAO+MoDTC oils.

Okubo et al. [113] also revealed the inhibition effects of ZDDP on the a-C:H wear in steel/a-C:H contacts under the lubrication of PAO+MoDTC+ZDDP. As shown in Figure 8a, under MoDTC, carburization of Mo occurred, which resulted in the formation of Mo-carbide-containing hard tribo-films. The hard tribo-films caused an abrasive wear of the a-C:H coating. However, under MoDTC+ZDDP, the additive-derived soft tribo-films inhibited both carburization and wear of the coatings.

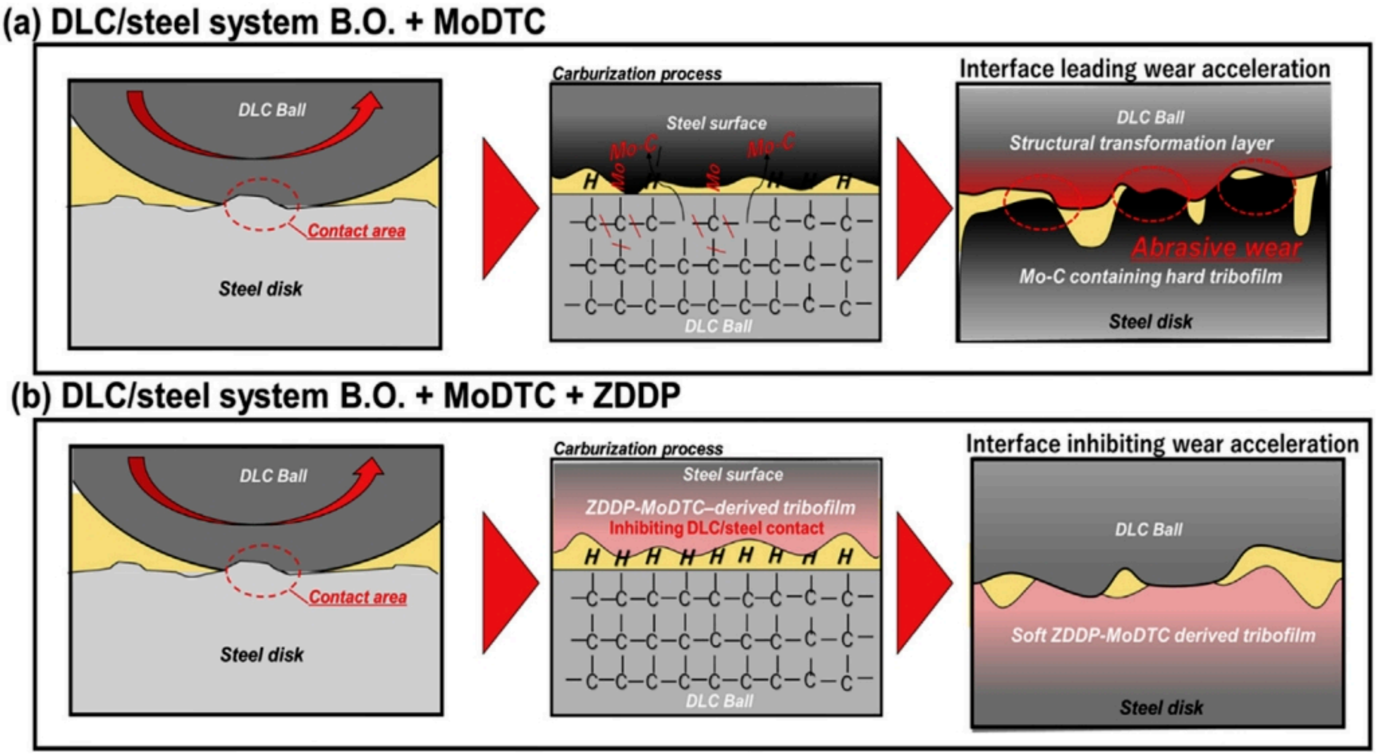


Figure 8. Schematic diagrams of the wear acceleration mechanism of a-C:H films lubricated with MoDTC solution and the inhibiting action of ZDDP on the wear acceleration [113].

2. Using DLC/DLC tribo-pairs to replace steel/DLC tribo-pairs

As the steel surface is essential for the wear acceleration of DLC coatings by MoDTC [117], one strategy to reduce the DLC wear in SLCL systems is using DLC/DLC tribo-pairs to replace the steel/DLC tribo-pairs. For example, the composite wear coefficients of a-C, a-C:H, Si-DLC, and ta-C:H self-mated tribo-pairs were extremely low under the lubrication of MoDTC-containing oils, while those of the corresponding DLC/steel tribo-pairs were very high, as reported by Vengudusamy et al. [111].

3.5.3. Synergistic Effects Between MoDTC and ZDDP in Oils

As MoDTC and ZDDP are the most widely used friction modifier and anti-wear agent, respectively, they are often used together in formulated oils [119]. Some reports found the synergistic effects between MoDTC and ZDDP on the tribological properties of oils due to the promoted formation of MoS_2. Consequently, the additive package of MoDTC+ZDDP was also used in the oils of some SLCL systems.

Kosarieh et al. [102] investigated the influence of MoDTC and ZDDP on the tribological properties of an SLCL system including a fully formulated oil and a tribocouple of 15 at.% hydrogenated DLC (a-C:15H) coating/cast iron. Under a low concentration of MoDTC (40 ppm Mo), the presence or absence of ZDDP in the oil had no influence on the COF and wear of the a-C:15H coating. A higher MoDTC concentration (300 ppm Mo) in the oil led to a lower COF due to the formation of MoS_2. However, a high MoDTC concentration resulted in a high wear of the a-C:15H coating due to the formation of MoO_3. Adding ZDDP into the oil with high-concentration MoDTC reduced the wear significantly due to the formation of glassy phosphate layers and the inhibited formation of MoO_3 particles.

The Liu group [120] measured the effects of MoDTC and mating pair on the tribological behaviors of two types of DLC coatings (a-C:H and ta-C) under high pressure (2.41 GPa) and elevated temperature (90 °C). For the a-C:H/steel pair, the addition of MoDTC to the base oil (API-III) reduced the COF from above 0.08 to around 0.05. However, for the a-C:H self-mated pair, the inclusion of MoDTC or MoDTC+ZDDP into the base oil increased the COF from 0.04 to 0.06. For both ta-C/steel and ta-C/ta-C pairs, MoDTC and MoDTC+ZDDP lowered the COF, and the oil containing individual MoDTC showed a lower COF compared with the oil containing MoDTC+ZDDP. The additive package of MoDTC+ZDDP reduced the coating wear significantly for all four pairs due to the formation of anti-wear tribo-films. Under the oils containing MoDTC or MoDTC+ZDDP, friction-reducing MoS_2 formed, as evidenced by Raman spectra, which well explained the reduction in COF upon the addition of MoDTC or MoDTC+ZDDP for the steel-mated and ta-C/ta-C pairs. However, some unexpected abrasive particles (Fe_2O_3, MoO_3, and β-$FeMoO_4$) were also generated in the presence of MoDTC+ZDDP, leading to the increase in COF in the a-C:H/a-C:H pair.

Haque et al. [121] compared the tribological behaviors of MoDTC and a Moly trimer in SLCL systems including a PAO+ZDDP oil. Compared with the oil PAO+ZDDP, the oil PAO+ZDDP+MoDTC and the oil PAO+ZDDP+Moly trimer produced a lower COF for three different tribo-pairs: a-C:H coating/cast iron, CrN coating/cast iron, and uncoated steel/cast iron. The a-C:H coating/cast iron and uncoated steel/cast iron pairs had a lower COF, while the CrN coating/cast iron pair showed a higher COF in the oil PAO+ZDDP+Moly trimer compared with that in the oil PAO+ZDDP+MoDTC. In particular, the a-C:H coating/cast iron tribo-pair showed a low COF at only ~0.05 in the oil of PAO+ZDDP+Moly trimer. Both MoS_2 and MoO_3 were detected in the tribo-films on the non-ferrous coatings under the lubrication of MoDTC or Moly trimer. And a higher MoS_2/MoO_3 ratio led to a lower COF. In addition, the surface graphitization of the a-C:H coating also contributed to the low COF.

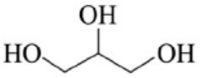
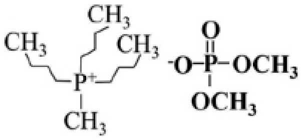
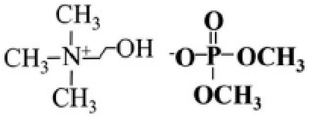
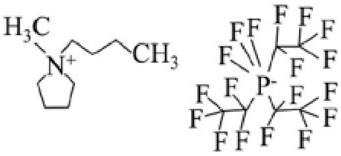
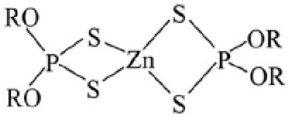
Besides ZDDP, some commonly used additives in engine oils can also reduce the wear of DLC coatings in the presence of MoDTC. Using steel ball-on-DLC-coated steel disc tests, The Ueda group [103] found the wear volume of the DLC wear track under PAO+MoDTC was above 3.5×10^4 μm^3, much higher than that under additive-free PAO (1×10^4 μm^3). The wear volume under PAO+MoDTC+Ca detergent, PAO+MoDTC+succinimide dispersant, or PAO+MoDTC+polyalkylmethacrylate was in the range $(0.5\sim1) \times 10^4$ μm^3. It was exciting that the wear volume under PAO+MoDTC+ZDDP, PAO+MoDTC+alkylated triphenyl phosphorothionate, PAO+MoDTC+overbased calcium salicylate, PAO+MoDTC+borated succinimide dispersant, or PAO+MoDTC+glycerol monooleate was very low (below 0.2×10^4 μm^3). These results indicated the DLC wear under PAO+MoDTC can be decreased greatly by adding some other surface-active additives. The authors suggested this might be because some anti-wear tribo-films formed between DLC and steel contacts, and the amount of MoO_3 derived from MoDTC was reduced by the competitive adsorption of the other additives. On the other hand, the COF of the steel/DLC pair under PAO was decreased by the addition of MoDTC, and all the two-additive systems (MoDTC and another additive) exhibited a lower average COF compared with the PAO+MoDTC system. In particular, the COF under the lubrication of PAO+MoDTC+glycerol monooleate was below 0.04 after a short running-in process. This investigation reveals that, when MoDTC is selected as the oil friction modifier in an SLCL system, an additive package including two or more additives rather than individual MoDTC is necessary for reducing both friction and wear of a steel/hydrogenated DLC pair.

3.5.4. Ionic Liquids (ILs)

ILs are molten salts at relatively low temperatures that are composed of ion pairs containing asymmetric cations and anions [122]. In the past two decades, ILs have attracted considerable attention in the field of lubrication due to their remarkable tribological behaviors, including excellent friction reduction, anti-wear performance, and high load-carrying capacity [123,124]. Like traditional ZDDP and MoDTC, the formation of protective tribo-films by tribochemical reactions is widely believed to be the origin of the superb tribological properties of ILs [125,126].

Some phosphorus-containing ILs have been considered as friction modifiers in oils because of the formation of phosphate boundary films during frictions [127,128]. Arshad et al. [129] investigated the effects of IL additives on the tribological properties of an SLCL system containing a base oil glycerol and a tribo-pair of steel ball vs. W-DLC coating. The information of the base oil, the IL additives, and ZDDP as a baseline comparison is listed in Table 2. Under 10 N and 100 °C, an ultra-low average steady-state COF of 0.024 was obtained for the lubricant G+AM or G+PP, lower than G and G+ZDDP. The formation of phosphate-based tribo-films played a significant role in the friction reduction. However, the IL-containing oils led to a higher wear of the steel balls compared with the ZDDP-containing oil. This may be attributed to the corrosion activities of the ILs towards the steel surface [130].

Table 2. Full names, abbreviations, and chemical structures of base oil glycerol, ILs, and traditional anti-wear ZDDP [129].

IUPAC Name	Abbr.	Mol. Structure
Base oil		
1. Glycerol, ($C_3H_8O_3$)	G	
Additives/ILs		
1. Tributylmethylphosphonium dimethylphosphate, ($C_{15}H_{36}O_4P_2$)	PP	
2. (2-hydroxyethyl) trimethylammonium dimethylphosphate, ($C_7H_{20}NO_5P$)	AM	
3. 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl) trifluorophosphate ([BMP][FAP]), ($C_{15}H_{20}F_{18}NP$)	BMP	
4. Zinc dialkyldithiophosphates, ($Zn[S_2P(OR)_2]_2$)	ZDDP	

In another report by Arshad et al. [131], they observed a 1.24 nm thick tribo-film on the W-DLC surface under the lubrication of a glycerol base oil containing a 1,3-dimethylimidazolium dimethylphosphate additive. This tribo-film involved dissociation of dimethylphosphate into dioxo-phosphate, which then interacted with W-clusters to form tungsten phosphates and cations. The reactions for the tribo-film formation can be summarized to seven steps, as shown in Figure 9. Step 1, sliding action generates a positive surface potential, and the anion part of the IL adsorbs onto the positively charged W-DLC surface, forming an electrical double layer. Step 2, the dissociation of dioxo-phosphate from dimethylphosphate occurs due to high temperature and mechanical load. Step 3, the dioxo-phosphate interacts with the W clusters on the surface of the W-DLC coating, forming tungsten phosphate. Step 4, tungsten phosphates are connected through oxygen bridging, forming phosphate-based layers. Step 5, the cations are attracted onto the phosphate-based layers. Step 6, prolonged sliding creates more and more patchy islands on the surface of the W-DLC coating. Step 7, the islands eventually combine and produce a continuous tribo-film.

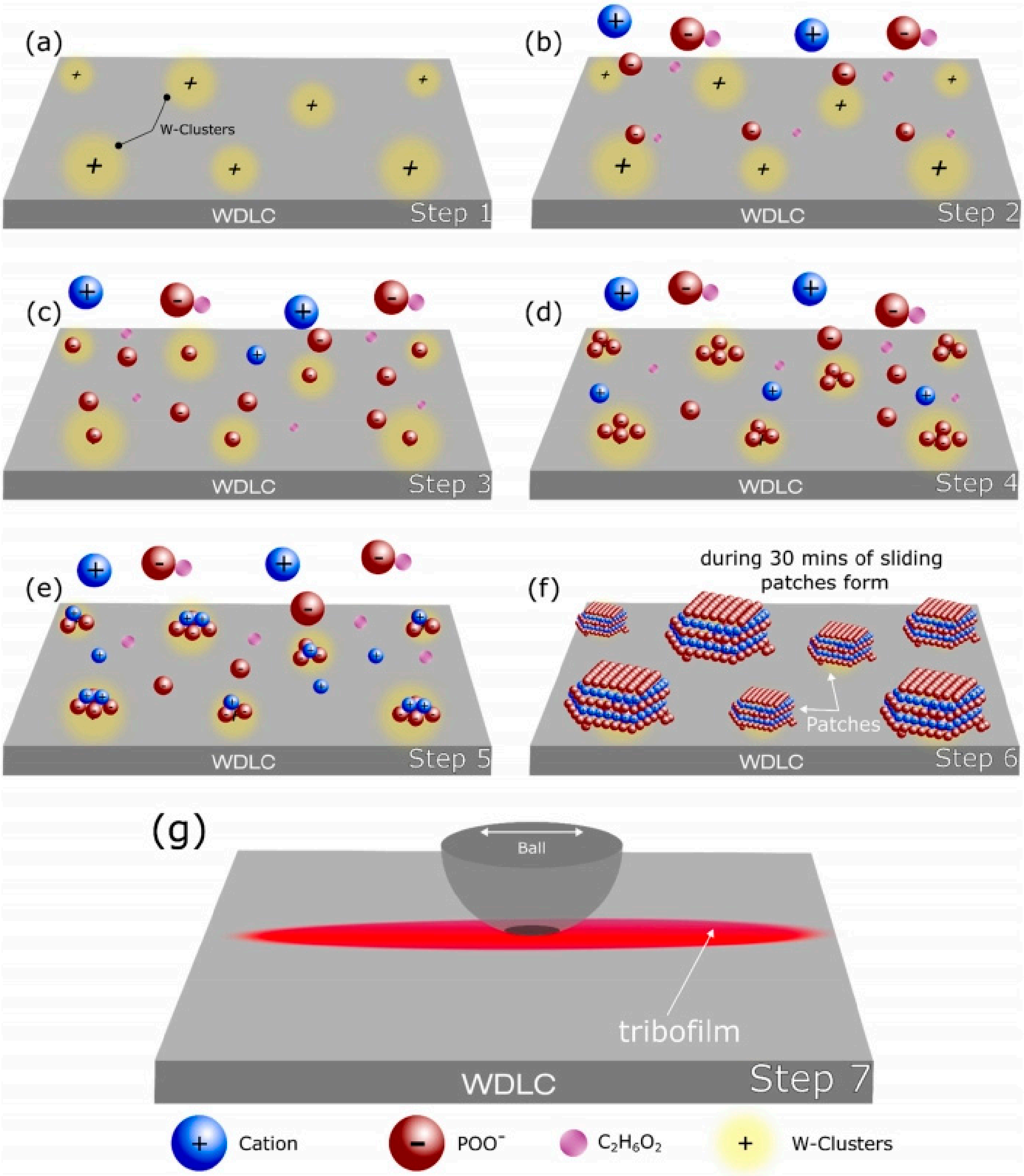


Figure 9. Schematic representation of interaction mechanism for IL on the W-DLC surface [131].

Qu et al. [132] compared the anti-wear properties of an ionic liquid, trihexyltetradecylphosphonium bis(2-ethylhexyl) phosphate ([P_{66614}][DEHP]), and ZDDP in a SAE 0W-30 base oil lubricating a-C:H coatings against steel balls. The tribo-film formed under [P_{66614}][DEHP] consisted of iron, metal oxides, and metal phosphates, and the tribo-film formed under ZDDP contained additional sulfides. The wear of steel balls under the IL was much less than that under ZDDP, indicating a superior wear protection of the IL to steel surfaces compared to ZDDP, in line with some previous reports [133,134]. For the a-C:H coatings, no measurable wear was observed under IL or ZDDP.

Based on the relevant reports, the friction-reduction and anti-wear mechanisms of ILs as additives in oils originate from the formation of tribo-films by tribo-chemical reactions, similar to the function of traditional anti-wear agent ZDDP. However, the corrosion activity of some ILs to steel needs to be avoided.

3.5.5. Other Oil-Soluble Additives

Commercial engine oils often include many kinds of oil-soluble additives. Consequently, the tribo-chemical reactions at the interface of tribo-pairs in SLCL systems with commercial oils are more complicated than the SLCL systems including a base oil with only one or two additives. The Espejo group [135] measured the tribological behaviors of three different tribo-pairs (steel/steel, steel/a-C:H, and steel/non-hydrogenated ta-C) by a pin-on-disc tribometer under the lubrication of a fully formulated oil containing some traditional additives (viscosity improver, ZDDP, antioxidant, detergent, and dispersant). It was revealed that no MoS_2 was formed when MoDTC was added into the fully formulated oil due to some unknown interactions. However, MoS_2 is often formed in SLCL systems with a simple oil consisting of a base oil+MoDTC+ZDDP [120,121].

3.6. Effects of Nanoparticle Additives in Oils

Recently, many kinds of nanoparticles (such as carbon [136], metals [137], and metal oxides [138,139]) are considered as solid additives for enhancing the friction-reduction, anti-wear, and load-carrying capacity of lubricating oils [140,141]. And the lubricants containing nanoadditives are called "nanolubricants" [142]. Two mechanisms, the ball bearing effect and the formation of low shear-strength tribo-film, are usually used to explain the working mechanisms of nanoparticles in the liquid nanolubricants. Recently, some nanolubricants have been tried in SLCL systems, and some representative research is presented as follows.

The addition of boron nitride (BN) nanoparticles to PAO base oil led to a significant COF reduction in an SLCL system to only 0.001 and a nonmeasurable wear [34]. This SLCL system contained a tribo-pair of Si_3N_4 ball vs. DLC film and PAO6 oil. The superlubricity was attributed to two reasons: firstly, nano BN behaved as nano-scale ball bearing to the pointlike contacts; secondly, the crystal layers in nano BN sheared easily and provided low frictions due to the weak van der Waals interaction forces between the layers.

Liu group [143] measured the effects of mono-dispersed Ni nanoparticles capped with oleylamine and oleic acid on an SLCL system containing a steel/DLC pair and di-iso-octyl sebacate (DIOS) oil. Experiments indicated the lubricating performance of the SLCL system was improved substantially after the addition of Ni nanoparticles, evidenced by a 50% reduction in wear rate of the DLC film and a 22% decrease in the average COF. The Ni nanoparticles facilitated the formation of tribo-films containing ferrous oxides, Ni, and its oxides.

Li group [144] investigated the influence of nano-oxides on the friction and wear behaviors of a non-hydrogen ta-C/steel pair under the lubrication of PAO. ZrO_2 nanoparticles reduced the friction but increased the wear of the ta-C coating; CeO_2 nanoparticles decreased the wear but increased the friction. The authors observed the formation of ZrO_2-condensed tribo-film, which was essential to the low friction and high wear of the coating. In the case of CeO_2 nanoparticles, the raised friction and inhibited coating wear were attributed to the existence of some wear debris (mainly iron oxides) and CeO_2 nanoparticles in the junction of the disc and cylinder's curve surface, which increased the contact area and reduced the contact pressure significantly.

In the Kassim group [72], Mo-containing particles (powder-type MoDTC, MoO_3, Mo_2C, MoS_2, and Mo) were added into PAO base oils separately in SLCL systems with a steel/DLC coating pair. Different from the other research aiming to reduce both friction and wear by particle additives, the purpose of this research is to identify which Mo-derived compound accelerates the wear of DLC coatings as all the tested particles are tribo-chemical products of MoDTC. Two types of DLC coatings were tested at room temperature: a-C:H and Si-doped a-C:H (Si-DLC). All the particles were negative to the friction reduction effect of PAO by increasing the average COF from ~0.05 of additive-free PAO to above 0.08 for particle-containing PAO. The highest average COF (above 0.16) occurred in the a-C:H coating under MoO_3-containing PAO, about twice that in the Si-DLC coating. The Mo_2C particles exhibited the most significant wear acceleration among the five types of additives, for both coatings. This may be explained by the high hardness of Mo_2C (15 GPa), making it an abrasive phase during friction. MoDTC powder-induced wear of the DLC coatings was almost negligible as the wear rates under pristine PAO and MoDTC powder-containing PAO were very similar. The MoO_3 particles, however, showed an anti-wear effect in PAO, though they were negative to the friction-reduction performance of the base oil. On the other hand, both MoS_2 and Mo particles contributed to a very low wear rate of the DLC coatings. Based on the experimental results, the authors suggested the Mo_2C particles were the major contributors to the severe wear of DLC coatings under the lubrication of MoDTC-containing oils. This research measured the tribological behaviors of the Mo-containing compounds separately; however, MoDTC-derived products by tribochemical reactions co-exist in most friction cases rather than exist individually, varied in their contents, components, and particle sizes due to changing working conditions, and MoDTC is usually used together with the traditional anti-wear agent ZDDP for obtaining a synergistic effect. Therefore, it may be more persuasive if an SLCL system containing all the MoDTC-derived products and ZDDP in the liquid lubricant was measured.

3.7. *Effects of Counterpart Materials*

In SLCL systems, the carbon diffusion performance of the counterpart material against DLC coatings is found to be an important factor that influences the wear of DLC coatings under oil lubrication. The diffusion or dissolution of carbon from DLC into steel occurred when the DLC coating was rubbed against steel, leading to a high wear of the coating. Aboua et al. [145,146] found that, under boundary base oil lubrication, the counterpart materials with lower carbon affinity/solubility resulted in lower wear of a-C:H coatings, compared with the counterpart materials with higher carbon affinity/solubility. Under PAO lubrication and elevated temperatures (80~120 °C), the a-C:H coating shows lower COF and wear against chromium (Cr) plating than against high carbon steel [145]. This is due to the formation of chromium oxide and chromium carbide layers on the surface of Cr plating, which can prevent the carbon diffusion. On the contrary, due to the high diffusion affinity of carbon atoms from coating to steel during friction, a high wear of a-C:H coating is induced when sliding against steel [147]. Through a comparison of the specific wear rate of a-C:H coating versus germanium (Ge), Cr plating, and S55C steel, a very low wear of the coating was obtained against Ge, by pin-on-disk tests at elevated temperatures (80~120 °C) under boundary base oil lubrication [146]. In addition, the pair of a-C:H coating vs. Ge disk had a low COF (0.04~0.06) in PAO at 120 °C. And the COF showed no significant variations when the normal load increased from 5 N to 30 N. The excellent tribological properties of the a-C:H coatings against Ge were attributed to the fact that there were no atomic interactions between the a-C:H coating and Ge counterpart, as evidenced by XPS characterizations. Ge is known to have extremely low carbon solubility [148]. Based on these observations, the authors deduced that the wear of a-C:H coating under boundary oil lubrication decreased with the carbon diffusion affinity of its counterpart material [149]. Therefore, counterpart materials with low carbon diffusion affinity (such as Ge and Ge-based surface coatings) should be selected in SLCL systems in order to achieve a low wear of DLC coatings.

4. Prospects

(1) One of the bottlenecks that seriously constrained the wide applications of SLCL systems is the high costs for depositing DLC coatings [150]. Based on the widely accepted deposition technologies, a whole set of equipment and the consumables (like the targets) for preparing DLC coatings are very expensive. Based on the markets, the costs of DLC coatings are more than ten times higher than commercial lubricating oils. And the DLC coating accounts for more than 90% of the cost of an SLCL system. Reduced costs of DLC coatings are essential for the commercial and engineering applications of SLCL systems. Therefore, relatively low-cost but efficient techniques need to be developed for fabricating DLC coatings. And the prepared DLC should have satisfactory tribological properties and excellent compatibility with traditional engine oils.

(2) Up to now, only some traditional particle additives have been used in the oils of the reported SLCL systems. Some new solid additives, such as black phosphorus [151],

MXene [152], carbon nanosheets [153], and some ball-like nano-hybrids [154,155], have not been tried in SLCL systems. Their compatibility and/or synergy with DLC coatings need to be investigated to construct efficient SLCL systems.

(3) The most widely used friction modifier in the oils of SLCL systems is sulfur-containing MoDTC. Some environmentally friendly sulfur- and phosphorus-free organic molybdenum (SPFMo) compounds have been synthesized and measured as new friction modifiers in lubricating oils in recent years [156,157]. However, there are few reports about SPFMo-containing SLCL systems.

(4) Doping of DLC coatings has been studied intensively and widely, as doping with additional elements can offset some drawbacks of the un-doped DLC. However, investigation about SLCL systems involving doped DLC coatings has been rarely reported.

(5) It is desired that an SLCL system can achieve superlubricity in practical engineering applications. Like DLC coatings and lubricating oils, optimized working conditions such as temperature, load, and velocity are essential for an SLCL system to realize superlubricity. In addition, the compatibility and synergy between the coating and the oil determine the tribological properties of SLCL systems. Consequently, the synergistic effects between coatings and oils need to be improved or optimized for the realization of the superlubricity of an SLCL system.

5. Conclusions

The newly developed SLCL systems are complicated, and the working mechanisms are also complex. However, some common observations were usually recorded in the previous reports:

(1) The DLC coatings often change their surfaces (such as graphitization) and/or participate in the tribo-chemical reactions during friction, both increasing the surface wear of the coatings.

(2) In a steel/DLC pair, the widely used additive package of MoDTC+ZDDP in lubricants usually results in a lower COF and wear compared with the lubricant without this package, due to the synergies between the formed friction-reduction MoS_2 and anti-wear tribo-films mainly derived from ZDDP [120]. However, for DLC self-mated pairs, this additive package usually led to an increased COF due to the generation of some abrasive particles.

(3) In the past two decades, many studies have reported the superlubricity of DLC films without the help of lubricating oils [158]. However, superlubricity in SLCL systems was rarely observed.

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