

Review



# **Recent Progress on the Tribology of Pure/Doped Diamond-like Carbon Coatings and Ionic Liquids**

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Abstract: This review provides a comprehensive overview of recent advances in tribology concerning pure/doped diamond-like carbon coatings (DLCs) and ionic liquid (ILs) interaction. DLC coatings are often used in industrial machinery and processes where sliding occurs between surfaces, leading to wear and degradation of their surfaces. DLC coatings are optimized by adjusting operating and deposition parameters as well as doping them with other elements to improve performance, such as thermal stability and chemical resistance. ILs are a promising green lubricant option due to their low melting temperature, superior thermal stability, and high miscibility with organic substances. ILs have been studied as neat lubricants and additives, and their tribological properties have been investigated, including their use as extreme temperature lubricants. The tribological properties of pure/doped DLC coatings with ILs have also been explored, although limited research has been conducted in this area. The combined effect of DLCs and ILs shows great promise in reducing energy loss due to friction, promoting longevity, and conserving energy.

Keywords: diamond-like carbon coating; doped-DLC; ionic liquid; tribology

# 1. Introduction

Concerns about energy and environmental sustainability have led to a need for more efficient and cleaner industrial activities and transportation systems. Fossil fuels are the main source of global energy consumption, which has a significant negative impact on the environment [1]. Vehicles alone consume about 30% of all energy produced [2], and a third of that is lost due to wear and friction. Friction losses and wear were identified as the primary culprits that restrict the lifespan and diminish the effectiveness of machinery used in the industry. As a result, these factors have a significant influence on the economy. This has a major impact on national economies, as shown by studies from several countries (United Kingdom [2], United States of America [3], Japan [4], and Canada [5]). Considerable resources have been dedicated globally to enhance the energy efficiency and wear resistance of moving mechanical parts. These endeavors, coupled with the increasing prevalence of electrically powered modes of transportation, represent a major stride towards meeting the demands for decreased carbon dioxide emissions set forth by the Kyoto [6], Paris [7], and EURO 7 [8] guidelines.

To decrease friction between sliding components and prevent mechanical wear, the conventional method is to utilize lubricants, typically mineral or synthetic oils in internal combustion engines or water-based emulsions in machining fluids. Additives are frequently incorporated into these fluids to enhance their existing properties or introduce new ones.



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**Copyright:** © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). Chemical compounds that can adsorb and/or interact with solid surfaces to generate low shear-strength reaction layers (known as "tribofilms"), are among the additives utilized to reduce friction (called "friction modifiers"), wear (known as "anti-wear additives") [9,10], and oxidation to modify rheology or even to add fragrances [11].

Zinc dialkyldithiophosphates (ZDDPs) have been utilized as additives in engine oil formulations to reduce wear since the 1940s. For many years, scientists have investigated the lubrication mechanism of ZDDP. The predominant view is that ZDDP functions as an anti-wear agent through a surface reaction that results in the formation of uneven phosphate films with a glass-like appearance. This theory has been widely accepted by researchers studying ZDDP's lubrication properties [12]. Although ZDDPs have proven to be highly effective in reducing wear across various conditions, their usage has also highlighted environmental concerns due to the significant levels of phosphorus, sulfur, and zinc they contain. These elements are recognized for their ability to cause clogging in filters and catalyst degradation in the exhaust after-treatment systems used in gasoline and diesel engines. As a result, the permissible levels of sulphated ash, phosphorus, and sulfur have gradually been restricted in modern engine lubricant formulations to address these environmental concerns.

Also, the oil-based lubricant constitutes an environmental threat. Moreover, the biodegradability of mineral oils is very low and the lubricant is released to the ambient environment in the form of microdroplets and accumulates on plants, animal, and ground-water tissues. The water pollution may be very aggressive since the oxygenation may be altered, leading to a disorder in the ecosystem. Soils also suffer serious damage from oil pollution due to the physical and chemical processes that lead to changes in the forms and distribution of organic matter and the range of carbon, water, nitrogen, and phosphorus, which can alter their entire ecosystem [11]. Because of all these concerns, researchers and companies have focused on developing natural-based oils to infer biodegradability in this product. Existing vegetable-based lubricants have good lubricity, ensuring a high cleaning effect. However, oxidative stability and low-temperature properties are their main drawbacks and require the integration of additives [11].

In recent years, there has been a growing interest in the use of ionic liquids (ILs) as a replacement or supplement to ZDDPs and other molecules in lubrication science and engineering, as well as in machining fluids. Ionic liquids are molten salts which consist of an organic cation and a weakly coordinating anion. They have become increasingly desirable in lubrication due to their exceptional properties, including high thermal stability [13], low flammability [14], and negligible vapor pressure [15]. Moreover, air-stable IIs with various structures and functionalities can be synthesized, making them a promising option for use in applications involving extreme conditions in space for components in space-shuttle [16], and micro-electromechanical systems [17,18]. As a result, IIs have garnered considerable attention as a potential solution for lubrication in such demanding situations.

Numerous studies have been conducted since 2001 when ILs were first suggested as potential lubricants [19–21]. In a recent study by Zhou et al. [22], it was demonstrated that phosphorus-based ILs react with cast iron surfaces and create phosphate-rich tribofilms that possess a similar chemical composition to those generated by other organophosphates, which are commonly used as anti-wear additives. Another area of research to reduce the use of lubricants has been the development of surfaces modified with self-lubricating coatings, such as diamond-like carbon (DLC) or transition metal dichalcogenides (TMDs). In the past few years, there has been significant interest in the research community towards combining diamond-like carbon (DLC) coatings with ILs due to their exceptional tribological properties such as resistance to wear and high levels of protection against corrosion thanks to their inert properties [23]. DLC coatings are composed of an amorphous network of carbon atoms that are in  $sp^3$  and  $sp^2$  hybridization and have hydrogen attached to terminate the dangling bonds [24–26]. The flowchart in Figure 1 explains the benefits of using DLC coatings along with ILs.



Figure 1. Tribological study of DLC coatings with ionic liquids lubricants.

This article presents a comprehensive overview of the current knowledge and advancements in the field of tribology of diamond-like carbon coatings and ionic liquids when used together. Firstly, the authors will describe DLC coatings concerning production and properties, and after that, the same will be done for ionic liquids. The tribology of DLC, undoped and doped, using ILs, will be discussed based on the literature results.

## 2. Diamond-like Carbon Coatings

# 2.1. Why DLC Coatings?

Industrial machines often operate in conditions where sliding occurs between surfaces (i.e., boundary lubrication regimes) and contact between asperities occurs, which leads to wear and degradation of components. Furthermore, in some industrial settings, there is a demand for sliding contact in the absence of liquid lubrication. To respond to this issue, selflubricating coatings have been developed to improve durability and performance in such situations that place high demands on the contacting surfaces. Self-lubricating coatings have been proven to provide decent outcomes in a wide range of technical applications. The application of these coatings modifies the microstructure, mechanical properties, and tribological performance of the contacting surfaces [27].

Self-lubricating coatings have a lower friction coefficient due to possessing a surface that can self-generate lubrication under certain conditions, such as heat or pressure. These coatings can improve the wear resistance of the material and extend the life of the parts they are applied to. However, achieving low friction and longer life simultaneously can be challenging. In some cases, increasing the wear resistance of the coating more prone to wear. Additionally, the conditions under which the coating operates can affect its performance, such as temperature, pressure, and environmental factors [27,28]. To overcome these challenges, researchers have been constantly developing new materials and coating that can provide both low friction and resistance to wear. This involves a careful balance between the material properties, deposition conditions, and techniques to optimize the performance of resultant coatings [28,29].

Diamond-like carbon (DLC) coatings are a type of self-lubricating coating known for their exceptional mechanical and tribological properties, such as high hardness, low friction, and excellent wear resistance [27,30]. Carbon coatings are classified as diamondlike carbon (DLC) or graphite-like carbon (GLC) based on the carbon fractions of  $sp^2$  and  $sp^3$  present in the coatings. In DLC coatings, the  $sp^3$  carbon fraction predominates, while in graphite-like carbon the  $sp^2$  fraction is the dominant one [31]. Also, a tribological response is heavily influenced by surface topology, as well as mechanical interlocking between surface asperities which leads to a higher coefficient of friction, particularly during the running-in stage. The roughness parameter of DLC coatings is largely determined by the topology of their substrate. For instance, achieving a low CoF is more challenging when applying this kind of film on rubber substrates [32].

DLC coatings come in a variety of types with different structural and chemical properties that can affect their tribological behavior. Some common types of DLC coatings include hydrogenated amorphous carbon (a-C:H), tetrahedral amorphous carbon (ta-C), amorphous carbon (a-C), and hydrogenated tetrahedral amorphous carbon (ta-C:H) films [33]. These coatings are used on parts like tappets, pistons, piston rings, fuel injectors, and biomedical devices (orthopedic, cardiovascular, and dental equipment), to lower friction. This reduction in friction not only improves savings but also aids in meeting regulatory bodies' stringent environmental and legislative requirements [33–35]. According to a report [36], the market size of diamond-like carbon (DLC) was assessed at USD 1786.04 million in 2021 and is anticipated to reach USD 3068.78 million by 2030. This growth is expected to occur at a Compound Annual Growth Rate (CAGR) of 6.30% during the period from 2023 to 2030. Moreover, the number of DLC publications indexed in the ScienceDirect database over the last ten years has increased from 550 publications published in 2012 to 1300 published articles only in 2022. The substantial increase in worldwide revenue and the corresponding rise in research output indicate the increasing significance and strong demand for DLC coatings.

#### 2.2. Major Research Trends in the Production Process of DLCs

Currently, there is an intense focus on enhancing and optimizing DLC coatings through two primary areas of investigation, first is operational and deposition parameters and the second is doping DLCs with other elements to improve their performance.

The first area pertains to operational parameters such as plasma potential, ion energies, and power source, which are being extensively analyzed for their impact on DLC quality. Physical vapor deposition (PVD) and plasma-enhanced chemical vapor deposition (PECVD) are the most common methods for the deposition of DLCs. Common technologies used for the deposition of DLC coatings are magnetron sputtering [37], filtered cathodic vacuum arc [38], microwave plasma [39], radio frequency (RF) glow [40], pulsed laser depositing [41], ion beam [42], plasma immersion ion implantation and deposition [43], and a combination of these techniques [44–46]. Each technology used for DLC deposition has multiple variables that affect its growth and performance, such as electric power inputs [47,48], distance factors (such as substrate to source distance) [49], and deposition temperatures [50,51]. Various methods, including sputtering, arc deposition, ion beam deposition, and hybrid methods (such as RF + PECVD), are being utilized to increase the  $sp^3$  ratio in DLC coatings [46]. It is worth noting that while a higher  $sp^3$  fraction leads to higher hardness, lower friction, and higher wear resistance, such coatings also have some drawbacks, such as higher residual stresses and lower toughness [46]. The aforementioned concerns have resulted in the expansion of the field of DLC structural design, encompassing monolayers [52–54], multilayers [55,56], and hybrid microstructures [46]. Kim et al. examined the impact of sliding velocity and temperature on the tribological properties of a multilayered DLC composed of Cr/CrN, a-C:H:W, and a-C:H layers deposited sequentially [57]. From this we can see that raising the speed resulted in a reduction in friction coefficients and the rates at which both surfaces experienced wear. This phenomenon was analyzed by conducting surface examinations. It was established that several factors contributed to the tribological characteristics, including the graphitization process and the formation of tribofilm on the worn ball specimens [57].

Data analytics-based optimizing techniques have been used to help researchers find the optimal parameter for deposition [58]. However, the best value for one parameter, such as bias voltage, does not necessarily result in the best combination of properties. Many parameter combinations may need refinement to achieve the best properties for DLC coatings. Jean et al. implemented the Taguchi method for optimizing deposition parameters and assessed the significance of process parameters by conducting an analysis of variance (ANOVA) [59]. In another study [60], various properties of DLC coatings, including their morphology, structure, bonding states, friction coefficient, and wear behaviors were optimized using an adaptive neuro-fuzzy inference system (ANFIS) identification technique assisted with Taguchi's algorithm to reduce the error of fuzzy estimator systems for tribological properties. Additionally, the optimal process parameters for the deposition of DLC films using plasma-enhanced chemical vapor process (deposition temperature, methane-argon flow rate, and hydrogen flow rate) were searched using the repulsive particle swarm optimization algorithm and a traditional genetic algorithm (GA) [61]. Solis-Romero et al. [62] optimized working settings for reducing the wear and friction of multi-layered a-C:H coating by implementing a hybrid grey-fuzzy algorithm. However, it is crucial to consider the fact that optimization or prediction works well when the model is made properly and accurately. A higher number of input parameters leads to more sophisticated models. Custom functions that are defined by the user have an impact on the results. Furthermore, the intricacy of the model has a bearing on its ability to be trained on the available data and also diminishes its potential to generalize well to new, unseen data [58].

The second area of investigation is related to the development of DLC nanocomposite coatings by introducing other elements. This approach involves doping the DLC coating with other elements (i.e., co-deposition of DLC coatings with metal, ceramic, and gaseous elements) to produce nanocomposites with improved properties [31]. For instance, to optimize tribological properties, DLCs have been doped with Cr [63], Mo [64], and Ni [65].

High residual stress in DLC films causes improper adhesion, which can lead to the peeling of coating even during film deposition. According to several studies [31,66–69], using metallic/non-metallic elements as dopants can reduce residual stress by controlling the microscopic structure, adhesion,  $sp^3/sp^2$  ratio, and surface roughness of DLC coatings. As a result, a controllable carbon bond structure and low residual stress are critical factors in obtaining preferable properties in a DLC film [68]. To reduce residual stress, DLCs have been doped with Ti [70,71], W [68,72], Si [73], N [74], F [69], Nb [67], Au [66].

Doping also improves the performance of DLC thin films in biomedical applications. This is the case for functional coatings in ureteral stents [75] or textiles for hospital use that need to be antibacterial [76], where, for example, Ag gives this property to DLC [77].

### 2.3. Characteristics of DLC Coatings

2.3.1. Thermal Stability of DLC Coatings

Another characteristic which researchers aim to enhance in DLC coatings is improving their maximum working temperature limit. This would allow DLC coatings to be useful in industries like aerospace, automotive, and turbomachinery, as they can currently only handle temperatures up to 300 °C [46,78]. When exposed to high temperatures, DLC films can experience various changes such as oxidation, graphitization, and dehydrogenation. Therefore, they can lose their lubricating capabilities and even detach from the surface they are applied to. This can lead to the creation of a transfer layer and wear debris on the surfaces of the objects in contact [79]. The graphitization phenomenon is thought to be responsible for lowering friction within the temperature range of 0 to  $150 \,^{\circ}$ C, in this range, heat can facilitate the transformation of some  $sp^3$  bonded carbons to  $sp^2$  carbons when a load is applied [80]. Similarly, the findings of Zhang et al. [81] showed that subjecting the DLC films to thermal annealing resulted in their graphitization. According to reports, elevating the annealing temperature caused a decrease in the hydrogen content in hydrogenated DLC films, which facilitated the shift from  $sp^3$  to  $sp^2$  hybridization. But they reported that at 300 °C and higher, the ultra-low friction behavior fails because the heat produced during friction might not dissipate promptly, and the system's temperature becomes too high to sustain a proper structure. However, introducing Si into the DLC films raised the  $sp^3/sp^2$  ratio, stabilizing the structure and preventing the films from graphitizing at high temperatures [79].

Wang et al. [82] reported on the volcano-type behavior of CoF in response to temperature increases in the 27 °C to 726 °C range for DLC thin films sliding against alumina and studied the mechanisms by implementing quantum chemical molecular dynamic simulations. CoF was stable in the 27 °C to 326 °C range, then after passing 326 °C, the CoF started to increase until it reached 527 °C, then again it began declining with increasing temperature up to 726 °C, which formed a volcano-type trend for CoF vs. temperature. At temperatures ranging from 326 °C to 527 °C, there is a formation of interfacial C-O and C-Al bonds between contacts, which results in a high friction coefficient. However, as the temperature passes 326 °C, the DLC undergoes graphitization, leading to a graphite-like surface that inhibits the formation of interfacial bonds and reduces the friction coefficient.

Yu et al. [83] studied the tribological characteristics of three different DLC films with varying Si contents applied on a tungsten interlayer. At temperatures below 300 °C, the pure DLC film exhibits impressive tribological characteristics. The low friction coefficient at room temperature is primarily due to the presence of Van deer Waals bonds, while the high-temperature properties between 100 °C and 300 °C are believed to be a result of graphitization. The addition of 14.56% Si content to the film allows for lubrication to be sustained at 400 °C, with the low friction coefficient obtained being a result of the enhanced thermal stability achieved through Si alloying. However, at temperatures higher than 500 °C, the film failed due to the oxidation of silicon. A composite film comprising oxidized tungsten and tungsten carbide phase is responsible for low CoF of Si-DLC with a tungsten interlayer.

### 2.3.2. Chemical Resistance of DLC Films

DLC's lack of reactivity with chemicals is highly desirable for its use as a shielding layer to prevent chemical reactions of the substrate with surrounding compounds [84,85]. To evaluate the chemical resistance of DLC films, the researchers polished aluminum substrates and applied DLC coatings of varying thicknesses. They found that thicker coatings offered more protection against chemical reactions than thinner ones, and that using a layered approach with a silicon interlayer was the most effective at preventing

permeation. However, the presence of defects in the coatings still had an impact on the permeation process [86]. In another study, Ohtake et al. conducted a concentrated nitric acid corrosion test to assess the acid resistance of the DLCs [87]. Although a high  $sp^3$  content was believed to reduce film corrosion, the actual relationship between  $sp^3$  content and corrosion resistance was found to be insignificant. Instead, corrosion resistance was found to be more closely linked to the presence of pinholes, micrometric growth defects, and dust particles. It was also discovered that increasing  $sp^3$  content and density during film application did not necessarily lead to improved corrosion resistance.

Wongpanya et al. [88] focused on examining the corrosion resistance, bonding structure, and biocompatibility of DLC films deposited on stainless steel with an interlayer of titanium. Results indicated that ta-C:Ti/Ti exhibited the highest level of corrosion resistance and biocompatibility, thanks to the formation of TiO<sub>2</sub> on its surface. Moreover, the presence of two barrier layers was observed in ta-C:Ti/Ti, making it a promising material for joint replacement.

Fayed et al. [89] examined the deposition of Si/DLC films on 2024-Al alloy using a plasma-enhanced chemical vapor method at different pulse voltages. Results showed that increasing the pulse voltage to 1800 V led to thicker coatings with better mechanical and corrosion resistance properties in 3.5 wt.% NaCl solution compared with other coatings due to an increase in  $sp^3$  hybrid in the coating, limiting electrical conductivity. However, increasing the pulse voltage to 2200 V increased the corrosion rate of the coatings.

## 2.3.3. Friction and Wear

Numerous research papers in the past few years have explored the frictional characteristics of DLC films in various settings [90]. The primary method by which DLC films typically control friction involves the formation of a transfer film, which then facilitates easy shear sliding within the interfacial material. However, the effectiveness of this mechanism is significantly influenced by the type of gas environment present in the contact area [91].

DLC coatings are highly susceptible to the effects of oxidizing agents, such as oxygen and water vapor, during friction. The presence of such agents can lead to significant tribo-oxidation of the topcoats, which in turn can increase both friction and wear. While wear rates can sometimes be very low, they can also be extremely high, rendering the coating unsuitable for certain applications [92]. According to Liu et al., the sustained low friction observed in DLC films under ambient air conditions is a result of wear-induced graphitization, which entails the creation of a tribolayer with a graphitic structure that exhibits low friction. The process of graphitization in their experiments was affected by two factors: the speed at which the surfaces were sliding against each other, and the amount of force being applied. This is because when the surfaces come into contact, the resulting friction causes the temperature to increase at the contact points. This rise in temperature causes hydrogen to be released from the DLC structure, which contributes to the graphitization process [93].

Films that have varying amounts of hydrogen possess distinct physical characteristics, including the way the carbon atoms are arranged, their refractive index, hardness, level of stress, and how they behave under controlled tribological conditions [91]. Miyake et al. conducted a study comparing the friction of two DLC coatings with different amounts of hydrogen. Both of the films had similar hardness values. The DLC film with a lower hydrogen content had higher friction in a vacuum environment than the film with more hydrogen. However, in humid ambient air, the DLC film with a lower hydrogen content had smaller friction compared to the film with a higher hydrogen content. The researchers found that the DLC film with a lower hydrogen content contained a mixture of diamond and graphite microcrystallites, resulting in friction behavior similar to that of diamond or graphite in a vacuum. On the other hand, no crystallized structure was observed in the DLC film with a higher hydrogen content. They speculated that hydrogen presence modifies the adsorption process of moisture and increases friction [94].

Donnet et al. reported that in PACVD systems, high-impact energy caused by bias directly controls the deposition process and leads to higher precursor dissociation. This results in a carbon network that is more crosslinked, has a lower  $sp^3$  fraction, lower hydrogen content, and a lower fraction of hydrogen bonded to carbon. Films deposited under such conditions are harder, have higher stress and index of refraction, lower surface energy, and exhibit very high friction (>0.5) in ultra-high vacuum (UHV) conditions. DLC films can achieve ultralow friction and wear in UHV if they have high enough hydrogen content (around 40 at. %), a crosslinked carbon network, and a noticeable fraction of unbounded hydrogen (about 0.3). By increasing the absolute bias from 500 to 800 V using the deposition system in the present study, the hydrogen content in films deposited from acetylene can be lowered by about 6 at. % and the fraction of hydrogen bonded to carbon by about 0.16, thereby increasing friction in UHV from less than 0.01 to more than 0.5. This indicates that the structure and composition of DLC films strongly affect their friction behavior, making it crucial to control both friction and wear by paying close attention to the deposition process [91].

In a similar study, a ball-on-disc reciprocation was used to investigate how hydrogenated and hydrogen-free DLC coatings behave when sliding against  $Al_2O_3$  in both air and pure water [95]. Under solid lubrication conditions in an air environment, the wear rate of DLC coatings sliding against  $Al_2O_3$  balls was primarily influenced by the coating's hardness. The presence of hydrogen in the DLC coatings reduced their hardness and increases their wear. Conversely, in water lubrication conditions, the hydrogen in hydrogenated DLC coatings plays a crucial role in determining the wear rate. Saturated  $sp^3$ -CH bonds in hydrogenated DLC coatings exhibited the lowest friction and wear, despite having lower hardness. On the other hand, hydrogen-free DLC coatings without H termination may cause relatively high friction and wear in water environments [95].

In addition to the hydrogen content, relative humidity also affects the tribological behavior of DLC coatings. Kokaku et al. [96] studied the impact of being exposed to a highly humid atmosphere on the wear and friction properties of hard carbon films. The exposed sample showed an increase in friction coefficients. This increase in friction coefficients occurred due to the oxidation reaction on the surface of the carbon, leading to changes in its surface properties. The creation of a carboxylic acid soap was responsible for the formation of the friction layer. This was found to occur when oxidized DLC films reacted chemically with ferrous oxide [97].

Kim et al. demonstrated how different normal loads and velocities can cause variations in CoF and wear [98]. An increase in load results in a reduction in the graphitization temperature due to the elevated contact stress. During experiments, it was observed that under constant sliding velocity, as the normal load increased, the average wear rates of both DLC coatings and AISI 52100 steel balls decreased. However, the average wear volume of the two surfaces increased. Conversely, as sliding velocity increased under constant load, both the average wear rates and wear volumes of the two surfaces increased until reaching a maximum value, after which they decreased as the sliding velocity further increased. The process of graphitization can be facilitated by friction-induced annealing on local contact areas and the strain energy induced by sliding [99].

# 3. Ionic Liquids

The development of biodegradable, environmentally friendly lubricants with lower toxicity levels is imperative. Within the realm of green chemistry, effective lubrication decreases energy consumption in mechanical applications, thereby reducing energy loss due to friction. The utilization of quality lubricants also protects all materials from friction and wear while promoting longevity. Additionally, employing an exceptional lubrication system aid in conserving energy, reducing energy loss, and minimizing raw material usage [100]. Recently, ionic liquids (ILs) have emerged as promising lubricant options for various applications. ILs, liquid salts composed of organic substances, are characterized by

a low melting temperature, high combustible temperature, low vapor pressure, superior thermal stability, low volatility, and high miscibility with organic substances [100–104].

Ionic liquids are characterized as being in a liquid state at temperatures below any arbitrary threshold, though the temperature requirement is not crucial for categorizing a substance as an ionic liquid [105]. Ionic liquids are very ancient and were first reported by Gabreil in 1888. Ionic liquids are constituted by the combination of organic cations and inorganic or organic anions, and as a result, the physicochemical characteristics of these liquids can be modified by varying the combination of cations and anions. This allows for the design of customized ionic liquids that are tailored to meet specific requirements [106,107]. So, ionic liquids are liquids that are composed of only ions and no other molecular species are present, with certain exceptions [105].

The mechanism of ionic liquids can be understood by the below Figure 2. Initially, as a consequence of sliding, the emission of free electrons from the metal surface occurs, resulting in the generation of a positively charged surface that elicits electrostatic attraction towards the anion moieties [108]. The formation of anion–cation layer-by-layer structures on metal substrates by ionic liquids is acknowledged, wherein the anion moiety securely anchors itself to the surface [109]. The resultant layered configuration serves as a protective measure, mitigating friction and wear on the surface [110]. An alternative mechanism may involve the generation of a reactive tribofilm, a phenomenon extensively discussed in the existing literature [110,111]. Throughout the process, certain molecules of the ionic liquid have the propensity to undergo dissociation and subsequently react with the metal present in the substrate, leading to the establishment of a tribochemical film.



Figure 2. Protective film formation of ionic liquids.

#### 3.1. Characteristics

Ionic liquids are recently trending for their peculiar characteristics and being viewed as green lubricants. The majority of researched ionic liquid lubricants contain imidazolium cations and either hexafluorophosphate or tetrafluoroborate anions [109,112–125]. As ionic liquids can be tailormade, which means there are infinite variations, and their properties can vary significantly, the more common characteristics for which they are usually preferred are non-volatility, versatile solubility, and thermal stability [126].

One of the main characteristics of ionic liquids is that they are green and sustainable, but some might contain phosphate, which is already an environment-depleting element, so how it can be sustainable? To answer this question, phosphate is commonly employed as a lubricating oil additive [127–135]. The creation of an ionic liquid exhibiting superior lubrication and anti-wear characteristics while posing no corrosion issues to metallic friction pairs can be achieved by coordinating phosphate with cations that are resistant to hydrolysis. Drawing inspiration from this concept, a series of alkyl imidazolium cation and phosphate anion-based ionic liquids were synthesized, and their tribological properties as lubricants were thoroughly studied [136]. The other characteristics of ionic liquids which are getting traction in current times is their use as lubricant in electric motors, as they have a wide electrochemical window [137], conductivity [138,139] and, for some of them, the ability to modify surface tension through the formation of micelles [140]. The first two of these properties are of fundamental importance for the use of lubricants in electric motors, as they inhibit the build-up of electrical charges. The second is important because it makes them compatible with other additives in the lubricant. It also increases their cleaning effect against aggregates formed over time.

## 3.1.1. Cationic Structure and Anionic Structure

Ionic liquids are composed of anions (ions carrying negative charge) and cations (ions carrying positive charge) [141]. Within an ionic liquid, the cationic structure often incorporates organic compounds, including ammonium or imidazolium, accompanied by alkyl chains and diverse substituents [142,143]. As for the anionic structure, it can be either inorganic or organic, exhibiting distinct sizes, shapes, and functional groups [142]. The properties of the ionic liquid, such as viscosity, conductivity, and solubility, are determined by the specific combination of cations and anions. Through the design of the cationic and anionic structures, the properties of ionic liquids can be tailored for various applications [144]. Furthermore, long alkyl chain substituents increase the tribological efficiency of cationic and anionic ILs but decrease their thermal stability. Hydrophobic ILs typically exhibit better tribological performance compared to hydrophilic ILs [145].

## 3.1.2. Thermal Stability

Thermal stability is a pivotal characteristic that impedes the degradation of lubricants within the working temperature range. At elevated temperatures, the lubricants are susceptible to chemical degradation, leading to fluid evaporation and viscosity reduction [146,147].

To evaluate the thermal stability of ILs dissolved in oils under nitrogen, thermogravimetric analysis (TGA) has been conducted extensively. Air is a crucial and practical medium for assessing the lubricating efficacy of ILs, wherein oxidation is inevitable. Typically, ILs exhibit greater thermal stability relative to hydrocarbon oils that decompose around 250 °C. Previous studies have indicated that phosphonium–carboxylate and ammonium– phosphate ILs display inferior thermal stability when compared to phosphonium–phosphate ILs. Conversely, imidazolium- and pyridinium-based ILs exhibit remarkable thermal stability, even in the presence of cationic alkyl chains [148].

In contrast to mineral oil and water, ILs exhibit negligible evaporation [146,147]. The thermal stability of a highly promising candidate for combustion engine utilization, ionic liquid is notably exceptional, exhibiting minimal mass loss even at temperatures up to 225 °C. Thus, the performance of ILs is deemed to surpass that of high-performance motor oils with regards to thermal stability [21]. Thus, they are deemed environmentally benign compared to conventional lubricants.

### 3.1.3. Corrosion Behavior of Ionic Liquids

To be applied in industry, ionic liquids must fulfil several conditions and a thorough test to check its potential corrosion behaviour towards equipment materials. The deterioration of materials can jeopardize reliability, decrease productivity, trigger shutdowns of the system, decrease yields, taint manufactured goods, lead to overdesign expenses, and necessitate expensive maintenance procedures [149].

One of the first comprehensive studies on this issue for flow-induced localized corrosion (erosion corrosion) cases was performed using the rotating cage technique and results showed that the tosylate and dimethyl phosphate anions have detrimental impacts. When diluted or contaminated with water, numerous anions utilized in the production of ionic liquids could undergo hydrolysis, leading to the creation of acids such as sulfuric and phosphoric acid. This process leads to an acidic environment which causes corrosion [149]. However, the focus of this research did not include the examination of localized attacks or flow-related analyses of ionic liquids. These areas warrant further investigation [149].

In a similar study with rotating cage setup, a screening was conducted of various ILs with differing ion structures to evaluate their corrosiveness behavior. The outcome of the study reveals a significant dependence of the corrosion behavior on the particular chemical structure of the ionic combination. It was observed that the vast majority of the tested ILs displayed a low level of corrosiveness in interaction with steel, particularly stainless steel [21]. Corrosion inhibitors refer to sub-stances that are added to the operating fluid in minimal quantities and chemically or mechanically interact with the metal surface to impede any further metal degradation and reduce corrosion [150–153]. Several studies have investigated the potential of ILs as corrosion inhibitors [150,154,155].

Therefore, while ionic liquids (ILs) can act as corrosion inhibitors, they can also promote corrosion in some cases. The corrosive behavior of ILs can depend on various factors, including the chemical structure of the IL and the metal surface, as well as the environmental conditions. These studies suggest that ILs can potentially act as corrosion promoters depending on their chemical structure and the conditions in which they are used. Therefore, it is important to carefully consider the specific IL and conditions when using them in industrial applications.

### 3.2. Ionic Liquids in Tribology

### 3.2.1. Tribological Behavior of Ionic Lubricants

Ionic liquids (ILs) have been widely implemented in lubrication systems, either as an additive or a neat lubricant. Researchers have demonstrated the remarkable outcomes of IL-based lubricants due to their exceptional properties and the potential to customize or substitute for traditional lubricants, thereby achieving superior results or improved output performance. Furthermore, the ability to vary the combination of cations and anions confers a significant advantage in the formulation of ILs for specific engineering and manufacturing applications. The physical properties of ILs and their potential have been extensively studied, primarily in the context of anti-wear and lubrication properties. The collective performance of ionic liquids reveals that when bio-based oil is mixed with ILs, the minimum coefficient of friction is achieved. The enhanced tribological performance of oil in the presence of ILs is attributable to the dipolar structure of ionic liquids, which adsorbs on the interacting surfaces and pro-duces a lubrication film [156]. However, the use of ILs as lubricants raises concerns regarding thermal oxidation, toxicity, corrosion, oil miscibility, and cost. Recent re-search has focused on developing thermally stable, non-corrosive, and oil-soluble ILs, which has been the subject of much discussion among researchers. The primary re-search on ILs in lubrication has shifted from utilizing them as both lubricant additives and neat lubricants to developing halogen and phosphorusfree ionic liquids as energy-efficient and environmentally friendly lubricant additives for steel-based engineering surfaces. The focus is to establish the correlation between the anion structure and the tribophysical properties of ILs. Halogen-free ionic liquids, such as borate-based ionic liquids, are of particular importance for lubricant applications in the present and future.

## 3.2.2. Ionic Liquids as Main Lubricant

Numerous research studies have been conducted on the use of ionic liquids as main (neat) lubricants [157]. Specifically, the use of three types of ionic liquids, namely 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate, 1-butyl-1-methylpyrrolidinium

bis(trifluoromethylsulfonyl)imide and ethyl-dimethyl-2-methoxyethylammonium tris (pentafluoroethyl)trifluorophosphate, in steel-to-steel contact has been investigated [158]. The results obtained from the studies show that the ionic liquid [BMP][NTf<sub>2</sub>] exhibited the lowest friction coefficient during the tests conducted at 40 °C and MTM tests as well as the lubricated surfaces with [BMP][NTf<sub>2</sub>] showed the lowest wear volume during tests conducted at 40 °C [158]. In another study, the effect of alkyl chain components on the chemical changes of various types of methylimidazolium salt was studied [143,159,160]. The presence of tribofilms in samples lubricated was confirmed by X-ray photoelectron spectroscopy, while only neat ionic liquid without interaction with the surface could be detected inside the wear scar of the sample lubricated, which suggests their usability as straight and neat lubricants. Figure 3 shows some of the ionic liquids which are used with DLC coatings as straight, additives and as both.



**Figure 3.** Some of the ILs which are used with DLC coatings as straight (main lubricant), additives and as both.

#### 3.2.3. Ionic Liquids as an Additive

The use of ionic liquids as lubricants presents a promising option for improving the cost-effectiveness of traditional lubricant systems. While the high cost of ionic liquids currently limits their use as neat lubricants to specialized applications, their incorporation as additives in small quantities in the base oils has gained interest due to their potential to enhance tribological properties [161]. By this it will be a good starting point for the use of ionic liquids in lubricants until a commercially viable cheap solution is devised. Therefore, a continuous drive for improved friction and wear performance for common tribo-pairs, as well as the need to facilitate lubrication in difficult-to-lubricate systems, necessitates the development of additive-based lubrication systems.

In addition to lubrication applications, research has shown that the use of ILs as lubricant additives and corrosion inhibitors can effectively reduce friction and wear. ILs have also been found to form electric double layers in water, allowing for the effective adsorption of additives on ceramic surfaces and improving tribological properties [162]. However, the use of water-based lubricants is limited under elevated test temperatures due to their high volatility and low cooling point, and the development of tribofilms on ceramic substrates is often thin. Compared to ILs, water-based lubricants may exhibit higher coefficients of friction and greater attachment of surface-clean alkyl phosphate [161].

## 3.2.4. Ionic Liquids as Extreme Temperature Lubricants

Ionic liquids can be used as lubricants in extreme temperatures. Researchers have studied the surface properties of metals lubricated with ILs at 300 °C. They found that ILs react with steel/iron substrates. New types of ILs have been developed, such as those with polyethylene glycol and functionalized dicationic, which exhibit excellent thermal stability and are reliable lubricants [163]. Some ILs, like imidazolium-based dynamic liquids, have high degradation temperatures which is greater than 400 °C, making them suitable for extreme temperature lubrication [164]. There are also different types of ionic fluid, such as polyacylater and polyifuoracillil, which have bridging moieties and can be tailored to have specific lubrication properties [164]. Thus, ionic liquids have a characteristic to maintain their properties at extreme temperature and pressure which makes them suitable for such conditions.

## 4. Tribological Testing of DLC Coatings with Ionic Liquid Lubricants

Tribological tests are designed to evaluate the friction, wear, and lubrication properties of materials in relative motion. In Figure 4, we can see the setups that help researchers and engineers understand the tribological performance of materials and lubricants, guiding the selection and design of components for various applications.

#### 4.1. Tribology of Pure Diamond-like Carbon Coatings and Ionic Liquids

Industry has long used solid lubricants to lessen wear and friction under varied circumstances. Advanced vacuum methods have been used to deposit materials like tungsten disulfide (WS<sub>2</sub>), hexagonal boron nitride (HBN), borides (MgB<sub>2</sub> and ZnB<sub>2</sub>), and soft metals like Cu, Ag, Sn, and Au as protective coatings or as lubricant additives. These substances provide a barrier that shields friction pairs from one another, reducing friction and enhancing wear resistance [165]. Because of its outstanding qualities, such as high hardness, excellent chemical stability, high thermal conductivity, low friction, and exceptional wear resistance, carbon has attracted the most interest of these solid lubricants [20]. There are various forms of carbon, and each form's characteristics rely on its particular structure. Researchers have intensively investigated different carbon forms and their applications over many decades [165]. Two specific forms of carbon, namely  $sp^3$  and  $sp^2$  form diamond-like carbon (DLC) thin films [165].



Figure 4. Testing process for DLC coatings with ionic liquid lubricants.

While operating under boundary lubrication conditions, applying solid lubricant thin films (coatings) can improve the lubrication of moving pairs [20,166]. A solid-liquid composite lubricating system can have a synergistic impact by integrating the benefits of liquid and solid lubrication. Solid lubrication coating has favorable load-bearing capabilities and low volatility. By doing so, the system is able to retain the benefits of solid lubrication while simultaneously gaining the benefits of liquid lubrication [167]. When the thin layers of fluid that separate surfaces fail, solid coatings can bear the load and serve as a secondary form of lubrication. This provides a backup mechanism to prevent direct contact between surfaces and reduce friction [168]. The combination of solid and liquid lubrication systems has become a popular choice for dynamic equipment, including space and automotive mechanisms. This composite lubricating system offers an effective solution for reducing friction and preventing surface wear. Thus, recent research has explored the potential benefits of integrating ionic liquids (ILs) with solid lubricating films. This promising approach has the potential to further improve the performance and durability of the composite lubricating systems [20].

Zhao et al. [169] examined the adhesion and friction of DLC and DLC coating in the presence of 1-octyl-2,3-Dimethylimidazolium bis(trifluoromethyl)sulfonyl ionic liquid as lubricant. To do so, a colloidal probe mounted on an AFM cantilever was used in contact mode to evaluate adhesive and nanotribological behaviors, and a UMT-3 tribometer was used in a ball-on-plate reciprocating mode to evaluate their microtribological behaviors. The experimental findings indicated that the friction forces of the DLC films with microgrooves were effectively reduced by introducing IL to the tribological pair. The reduced adhesion and friction forces were explained by lubricity property of ionic liquid to prevent

direct contact between the DLC coatings and colloidal tip, which made the sliding of the colloidal tip on the DLC coating's surface easier. The reciprocating test setup involves applying back-and-forth sliding motion to examine the wear and friction characteristics of materials, particularly in reciprocating applications such as ball-on-plate reciprocating mode or a piston-cylinder system. This setup proves valuable in assessing the performance and durability of components subjected to reciprocating motion which is shown in the below Figure 5.



Figure 5. Pin-on-plate reciprocating tribological test setup.

The outcomes of the experiment that tested the friction coefficient of three PVD coatings using [BMP][FAP] lubricant demonstrated comparable values for TiN, CrN, and DLC, with CoF of DLC being the smallest, which suggests that the ionic liquid had a noticeable impact on the coatings regardless of their type [170]. To gain a more profound understanding of the interactions between the coatings and the ionic liquid at a chemical level, XPS analyses were conducted to examine the wear surfaces. The breakdown of ILs starts the process, and the dynamic components in the ILs may respond with the newly formed surface to create a reaction film, also known as a tribofilm. This tribofilm is capable of protecting the surface from significant wear. A layer is formed on the surface of DLC by the ionic liquid, where 40% of the fluorine is involved. When the experimental conditions became harsher, causing an increase in temperature and pressure, the interaction rate also raised. As a result, roughly 80% of the overall fluorine became connected to the surface within the wear scar, in contrast to 40% outside the scar [170]. Another study investigated the same coatings but in the presence of another ionic liquid described as [(NEMM)MOE][FAP]. When the films were lubricated with pure ionic liquid ([(NEMM)MOE][FAP]), the lowest friction coefficient was achieved compared to when used as an additive to PAO 6 as base oil, and no noticeable wear scar was identified. The behavior of the ionic liquid and DLC interaction differed significantly from that observed in other films. The ionic liquid does not interact with the surface of DLC unless there is a significant increase in loading conditions. As a result of significant increase in loading conditions, approximately 77% of the fluorine (F) reacts with elements inside the wear region. When compared to previous research conducted under identical testing conditions, the [BMP][FAP] ionic liquid displayed marginally superior performance as a pure lubricant and as an additive in the lubrication of the three PVD coatings examined, relative to the [(NEMM)MOE][FAP] ionic liquid. The improved tribological behavior of [BMP][FAP] can be attributed to its higher viscosity and stronger interaction with these coatings, which encourage the growth of tribofilms [170,171].

Jia et al. examined the effectiveness of synthetic ionic liquid functionalized borate esters as additives in PAO as the base lubricant in terms of friction and wear properties for DALC films compared to ZDDP [172]. According to the results obtained, the addition

of borate esters in PAO provided improved friction and wear performance for DLC films in comparison to ZDDP. The authors hypothesized that a triboplasma forms during the process, particularly on DLC films, and that the borate esters modified with ionic liquid might adhere to the worn surface of the pair because of the triboplasma. Furthermore, the decomposition of the borate esters during the process leads to the B atoms infiltration into the flaws and sublayer of the worn surface. Consequently, a tribofilm containing B, N, and F was formed to lower friction and reduce wear [172]. Table 1 shows the brief summary of the undoped diamond-like carbon coatings and ionic liquids.

**Table 1.** Summary of the available literature investigating tribological performance of undoped diamond-like carbon coatings and ionic liquids.

Name of ILs	Tribometer	Additive or Main Lubricant?	Ref.
1-octyl-2,3-Dimethylimidazolium bis(trifluoromethyl)sulfonyl	UMT-3 tribometer (ball-on-plate)	Used as main lubricant (100 wt.%)	[169]
1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate	CETR-UMT-3 micro-tribometer (ball-on-plate)	Used as both additive in PAO 6 (1 wt.% additive) and main lubricant (for comparison)	[170]
ethyl-dimethyl-2- methoxyethylammonium tris(pentafluoroethyl)trifluorophosphate	UMT-3 microtribometer (ball-on-plate)	Used as both additive in PAO 6 (1 wt.%) and main lubricant (for comparison)	[171]
The synthesized ionic liquids of functionalized borate esters (The specific names are not mentioned but molecular structure is explained in the article)	Reciprocating friction and wear tester (ball-on-disk)	Used as 2 wt.% additives to polyalphaolefin (PAO 6) and compared with 2 wt.% ZDDP as a reference additive	[172]
(1) 1-butyl-3-methylimidazolium tetrafluoroborate And (2) trihexyltetradecylphosphonium bis(trifluoromethy-lsulphonyl) amide	T-01M tester (ball-on-disc configuration)	Used as main lubricant (100 wt.%)	[173]

Another study investigated the performance of two distinct ionic liquids in conjunction with a friction pair comprised of a steel ball and a steel disc coated with DLC. In this comparison, the 1-butyl-3-methylimidazolium tetrafluoroborate ionic liquid outshone its counterpart, the trihexyltetradecylphosphonium bis(trifluoromethylsulphonyl) amide ionic liquid [173]. Notably, the former demonstrated superior efficacy in lubricated friction and exhibited compatibility with a-C:H type DLC coatings. The friction pair, consisting of a steel ball and a steel disc with DLC coating, played a crucial role in the evaluation. The steel ball, a standard material for friction testing, offered durability and widespread applicability in industrial settings. Meanwhile, the steel disc's DLC coating contributed to enhanced wear resistance and served as a protective layer, prolonging the material's lifespan. Exploring possibilities for synthesizing or mixing these ionic liquids could further optimize their performance. Such endeavours may lead to improved lubrication properties, enhanced compatibility with DLC coatings, and potentially cost-effective solutions. The ball-on-disk test setup, depicted in Figure 6, served as a valuable tool by allowing the examination of contact between the ball and rotating disk, providing insights into friction and wear properties. This testing methodology finds extensive applications in both sliding and rolling conditions, offering a comprehensive assessment of material performance and durability in various industrial contexts.





# 4.2. Tribology of Doped Diamond-like Carbon Coatings and Ionic Liquids

In their investigation, Madej et al. [174] explored the tribological properties of diamondlike carbon (DLC) coatings, specifically a-C:H and a-C:H:W, fabricated using plasmaassisted chemical vapor deposition (PACVD) and physical vapor deposition (PVD) methods. These superhard anti-wear coatings were applied to steel elements and subjected to scrutiny under both dry and ionic liquid lubricated conditions. The study utilized atomic force microscopy (AFM) and scanning electron microscopy (SEM) for comprehensive surface analysis, examining topography and cross-sections. Tribological assessments, employing a ball-on-disc tribometer and a pin-on-plate tribometer, revealed that the DLC coatings outperformed the steel substrate, displaying lower wear, reduced friction, and higher hardness [174]. To delve further into the wear resistance and tribological performance, there is potential in exploring synthesis possibilities such as optimizing blending ratios or combining PACVD and PVD methods. The pin-on-disk test setup, elucidated in Figure 7, emerged as a pivotal tool in assessing friction and wear characteristics in sliding contact applications. This comprehensive study contributes valuable insights into the advanced properties of DLC coatings. Table 2 shows the brief summary of the doped diamond-like carbon coatings and ionic liquids.



Figure 7. Pin-on-disk tribological test setup.

Name of Ionic Liquids	Tribometer	Doped DLC Type	Ref.
dialkyloimidazolium tetrafluoroborates (Used as main lubricant, i.e., 100 wt.%)	T-01M tribometer (ball-on-disc configuration)	W-doped DLC	[174]
tributylmethylphosphonium dimethylphosphate (PP) (used as additive with 1 wt.%) and 1,3-dimethylimidazolium dimethylphosphate (IM) (used as additive with 1 wt.%) and 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl)trifluorophosphate	TE 38-Phoenix Tribometer (reciprocating pin-on-disc tribometer)	W-doped DLC, and Ag-doped DLC	[23]
1,3-dimethylimidazolium dimethylphosphate (used as additive with 1 wt.%)	UMT-2 tribometer with (ball-on-flat-disc geometry)	W-doped DLC	[175]
<ul> <li>(1) 1-butyl-1-methylpyrrolidinium tris(pentafluoroethyl) trifluorophosphate ([BMP][FAP]) (used as additive with 1 wt.%) and</li> <li>(2) tributyl-methyl-phosphonium dimethylphosphate (PP) (used as additive with 1 wt.%) and</li> <li>(3) (2-hydroxyethyl) trimethylammonium dimethylphosphate (AM) (used as additive with 1 wt.%)</li> </ul>	UMT-2 tribometer (reciprocating motion with a ball-on-flat-disc geometry)	W-doped DLC	[176]
(1) tetrafluoroborate (LB104) or [BF4] (LB104) (used as main lubricant, i.e., 100 wt.%) And (2) tetrafluoroborate (LAB103) or [BF4] (LAB103) (used as main lubricant, i.e., 100 wt.%)	CSM Switzerland tribometer (ball-on-disc)	Cr-doped GLC and Cr-doped DLC	[26]
trihexyltetradecylphosphonium bis (2-ethylhexyl) phosphate [P_66614] [DEHP] (used as additive with 1 wt.%)	Block-on-Ring configuration	Gd-DLC and Eu-DLC	[177]
trihexyltetradecylphosphonium bis(trifluoromethylsulfonyl) amide (used as main lubricant, i.e., 100 wt.%)	Anton Paar TRB tribometer (Ball-on-disc configuration)	Si-DLC	[178]
1-alkyl-3-octylimidazolium hexafluorophosphate (L-P801) (used as main lubricant, i.e., 100 wt.%) And 1-alkyl-3-octylimidazolium hexafluorophosphate (L-P804) (used as main lubricant, i.e., 100 wt.%)	UMT-2MT (reciprocating ball-on-disk)	Ti-DLC	[179]

**Table 2.** Summary of the available literature investigating tribological performance of doped diamond-like carbon coatings and ionic liquids.

Hamid K. et al. studied tribological performance of three DLC coatings (DLC, Wdoped DLC, and Ag-doped DLC) deposited on stainless steel substrates in dry and lubricated conditions using a water-based main lubricant additivated with three ILs [23]. The doped coatings demonstrated superior mechanical integrity, toughness, and adhesion compared to undoped DLC. Among the coatings, the Ag-doped DLC had the best mechanical properties. Tungsten carbide precipitates were formed by W in the DLC coating. Friction was controlled by two different additive-adsorption mechanisms: a triboelectrochemical activation mechanism for Ag-DLC and an electron-transfer mechanism for W-DLC, resulting in the largest friction reduction [23]. The friction of DLC (diamond-like carbon) is influenced by the electrical charge of the surface and the lubricant's ability to adsorb to the surface. The electrical conductivity of the lubricant (ionic liquid) drives the transport of additives to the surface and impacts friction. The higher the lubricant's electrical conductivity, the lower the friction. Additionally, the Zeta-potential of DLC surfaces in aqueous solutions is negative, resulting in no change in friction with the addition of negatively charged molecules [180,181]. The higher electrical conductivity in IL-additivated lubricants leads to faster transport kinetics of the cation and anion moieties to the surface, resulting in lower friction [23].

Arshad M. et al. explored the potential of enhancing the lubrication properties of tungsten-doped diamond-like carbon coatings by incorporating a 1,3-dimethylimidazolium dimethylphosphate ionic liquid into glycerol base oil. The researchers conducted tribological tests under varying loads (5 N, 10 N, 20 N) and elevated temperature (100 °C). They found that the friction coefficient was significantly reduced by approximately 50% when 1 wt% IL was included. They also discovered that the formation of a thin tribofilm on the surface of the coatings played a crucial role in friction reduction. The study provides insights into the lubrication mechanism and offers implications for the development of improved lubricants for tribological systems [175].

Another investigation was conducted by Arshad M. et al. to examine the interaction between coatings of tungsten-doped diamond-like carbon (WDLC) and three phosphate-based ionic liquid (IL) additives. Among these additives, two contained the anion dimethylphosphate, while the third contained the hydrolytic trifluorophosphate anion. The tests were carried out under boundary-lubrication conditions. The findings indicated that IL additives containing dimethylphosphate anions exhibited reduced friction on the surface of WDLC, whereas the IL with trifluorophosphate anion displayed poor performance. Analysis of the surface revealed the formation of a tribofilm based on phosphate on the WDLC surface when dimethylphosphate additives were present, resulting in friction reduction. The WDLC coatings demonstrated remarkable resistance to wear [176].

Researchers conducted a study on Chromium doped diamond-like carbon coatings (Cr-DLC) using PVD and PECVD methods [26]. The lubrication performance of solid-liquid composite lubrication systems was studied using two ionic liquids (ILs) as lubricants. The findings indicated that the friction coefficient was reduced by about 40% when compared to dry conditions, and the composite system displayed an effective synergistic lubrication effect. The Cr-DLC coating showed superior tribological, mainly because of its improved physicochemical film formation during friction and the dense microstructure of the Cr-DLC coating. The ILs' viscosity, corrosiveness, and coating microstructure influenced the composite systems' synergistic effect [26].

Rare earth metals, such as Gd and Eu, present an intriguing avenue for enhancing the reactivity of diamond-like carbon (DLC) when incorporated into its composition. Studies indicate that DLC films doped with low atomic concentrations (1–3 at.%) of Eu or Gd, particularly in dry contact conditions without lubrication, exhibit a marginally higher coefficient of friction (CoF). However, these films demonstrate commendably low wear rates and boast high hardness, rendering them well-suited for diverse applications [177]. Furthermore, investigations by Shaikh et al. [182] and Sadeghi et al. [183] highlight that DLC coatings doped with gadolinium can yield an even lower coefficient of friction compared to pure DLC when subjected to trihexyltetradecylphosphonium bis(2-ethylhexyl) phosphate [P<sub>66614</sub>] [DEHP] ionic liquid (1 wt.%) as an additive to polyalphaolefin (PAO) 8. This is due to the reactivity of the ILs and dopants, to explore the possibilities and advantages of synthesizing or mixing these materials, attention should be given to optimizing the concentration of rare earth metal dopants for optimal tribological performance. The

Block on Ring test setup, illustrated in Figure 8, played a central role in these investigations. This setup involves pressing a stationary block against a rotating ring, enabling the measurement of friction and wear characteristics between the two components. Widely applied in assessing materials for brake linings and clutch facings, the Block on Ring test provides valuable insights into the durability and tribological properties of materials under conditions resembling real-world applications. After the tests, the results were confirmed using the calculations in the stribeck curve and SEM/EDS analysis which showed presence of phosphoros from ionic liquid on the surface of the doped DLC coating.



Figure 8. Block-on-ring tribological test setup.

Similar study examined the effect of a silicon doping on tribological behavior of diamond-like carbon coating in IL-lubricated friction pairs [178]. Tests showed that the Si-DLC coating, when used with the ionic liquid, reduced both coefficient of friction and wear. The study concluded that using Si-DLC coatings lubricated with the ionic liquid helps to improve tribological properties of sliding surfaces under friction [178].

In another study, Ti-doped DLC coatings were analyzed using Raman and TEM. Two types of 1-alkyl-3-octylimidazolium hexafluorophosphate ILs (L-P801 and L-P804) were synthesized and evaluated as lubricants for Ti-DLC/steel contacts, with excellent friction-reducing properties. To investigate the chemical components of tribofilm formed on the contacting surface for Ti-DLC films in the presence of L-P801 ionic liquid, XPS characterization method was applied which confirmed the existence of PF6 anion, anime and/or nitrogen oxide, and fluoride on the surface. These coatings with ILs lubricating systems have potential as lubricants in vacuum and space moving friction pairs [179].

Limited research has been conducted on the tribological properties of doped DLC coatings with ionic liquids (ILs), and their combined effect has been found to be highly beneficial. The addition of doping elements has been demonstrated to improve the tribological properties of DLC coatings significantly. ILs, with their exceptional characteristics such as excellent lubricity and high thermal stability, are deemed ideal for various tribological applications. Through the formation of a protective film or tribofilm on the surface of the coating, the combination of doped DLC coatings and ILs has been reported to elevate their tribological properties further.

Ionic liquids (ILs) are increasingly used in tribology, particularly as lubricating agents, with extensive assessment in laboratory settings [100–104,106,107,184]. This review emphasizes the need for improved efficiency and environmentally friendly practices in industrial operations and transportation systems, addressing concerns about energy conservation and environmental sustainability. The exploration focuses on the application of diamond-like carbon (DLC) coatings and ionic liquids (ILs) in tribology, covering their inherent characteristics, testing, synthesis, and potential applications.

Preferably, the anionic constituents ought to manifest hydrophobic tendencies, thereby enhancing tribological attributes and thermo-oxidative stability. The incorporation of larger alkyl groups within imidazolium structures augments tribological performance but concurrently precipitates a decline in thermo-oxidative stability. The augmentation of anti-wear properties in ionic liquids can be achieved via additive technologies. Substantial room for optimization still exists, especially in the arena of mitigating friction and inhibiting corrosion. Notably, DLC coatings, particularly those imbued with doping agents, have emerged as a boon in this regard, as the coupling of doped DLC coatings with ILs has yielded notably diminished friction coefficients and reduced wear rates [182].

Given the recognition of ionic liquids as bespoke fluids, substantial endeavours must be undertaken to engineer innovative lubricants. Notably, halogen-free anions are in high demand. Cationic constituents exhibit a greater degree of flexibility in comparison to their anionic counterparts, and cations possessing multifunctional groups are of pronounced interest to the research community.

Doped DLC coatings synergize effectively with the pronouncedly polar nature of ILs, and this amalgamation of characteristics is beginning to be harnessed within the domain of tribology, endowing the field with advantages such as heightened load-carrying capacity, the formation of tribolayers subsequent to interaction with sliding surfaces, energy conservation through the mitigation of friction coefficients, and the reduction of wear rates for sliding materials. These advancements stand to render DLC coatings applicable across a spectrum of industries, encompassing aerospace, automotive, and turbomachinery.

The sheer diversity in the composition of anions and cations, lateral substituents, and self-organization within diverse base fluids necessitates a concerted effort to discern the optimal combination for each pragmatic application. ILs are characterized by their low melting points, elevated combustion thresholds, minimal vapor pressures, remarkable thermal stability, low volatility, and remarkable miscibility with organic substances. Within the ambit of this review, the tribological performance of ILs is dissected, encompassing their role both as standalone lubricants and as additives, their application within extreme temperature lubrication contexts, and their potential in boundary lubrication scenarios when paired with solid lubricating coatings.

For ILs to survive in the competitive atmosphere of lubrication and tribology, they must exhibit effectiveness not solely as autonomous lubricants but also as supplements to other fluids, including water, mineral oils, or synthetic oils. Ionic liquids find utility as lubricants in vacuum machinery within space applications. Moreover, their application extends to high-temperature environments characterized by substantial fire risks. Lubricants boasting exceedingly low vapor pressures emerge as highly desirable for machinery deployed within clean rooms where hazardous gases are stringently regulated.

The amalgamation of Ionic Liquids (ILs) with Diamond-Like Carbon (DLC) coatings in emerging technologies exhibits considerable promise, albeit accompanied by intricate challenges. Notably, the stability and compatibility of ILs with DLC coatings present formidable hurdles, necessitating sustained efficacy under diverse operational conditions. The synthesis complexity inherent in the fabrication of DLC coatings, coupled with the nuanced integration of specific ILs, mandates sophisticated manufacturing methodologies to ensure uniform deposition and controlled application [182]. Furthermore, environmental ramifications, particularly concerning the potential toxicity or ecological impact of certain ILs, underscore the imperative to identify environmentally benign alternatives.

In terms of extensible dimensions and prospects, the amalgamation of ILs with DLC coatings delineates a trajectory towards advanced tribological solutions. This innovation holds promise for the development of sophisticated lubrication systems applicable in high-performance machinery, automotive engines, and aerospace components. Although the production costs associated with certain IL families, such as imidazolium derivatives, continue to present a challenge in terms of competitiveness with established commercial lubricant additives, the emergence of novel ILs via straightforward synthetic routes is noteworthy. Furthermore, the utilization of ionic liquids as additives within synthetic and mineral oils has yielded fruitful outcomes, culminating in the reduction of friction coefficients and minimal wear. This effect is further accentuated when combined with doped DLC coatings [182]. Ongoing research endeavours offer prospects for tailored tribological solutions, wherein the customization of IL-DLC technology aligns with specific industrial requisites. Notably, the judicious selection of environmentally friendly ILs contributes to the overarching goal of environmental sustainability, aligning with global initiatives for greener industrial practices. In summation, while challenges persist, the extensible dimensions and prospects of Ionic Liquids with DLC coatings underscore their potential as a groundbreaking tribological solution, necessitating concerted efforts to surmount existing impediments and harness the envisioned advancements in diverse scientific and industrial domains.

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