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Tribological properties of hydrogen doped ta-C coating under high temperature and high vacuum

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1. Introduction

In recent decades, sliding machines have been increasingly required to operate in various extreme environments, such as high vacuum, high speeds, heavy loads, and super low or high temperatures. Notably, in high-temperature environments, numerous moving machine assemblies—including those used in power generation, aerospace, petrochemical industries, metalworking, and other industrial sectors—are subjected to these conditions. Friction and wear are critical factors in determining the efficiency, durability, reliability, and robustness of tribological systems in such applications. High-temperature friction processes are highly complex, and the need for both fundamental and applied research in this area is becoming more urgent.

To achieve low friction and high wear resistance in sliding surfaces for industrial applications, Diamond-like Carbon (DLC) coatings have garnered significant attention. Among the various types of DLC, tetrahedral amorphous carbon (ta-C), characterized by a high fraction of sp^3-hybridized carbon, is considered an effective low-friction, high wear-resistant coating under oil lubrication. ta-C coatings have been successfully applied to automobile components, reducing friction in engine oil, including those containing friction modifiers like MoDTC [1]. Additionally, nitrogen-doped ta-C, known as ta-CNx, has demonstrated lower friction and higher wear resistance in environments such as base oils, vacuum, and dry nitrogen gas [2-5].

However, ta-C has exhibited high friction in high-vacuum conditions, as observed by Fontaine et al. [6], which demonstrated the super-low friction properties of hydrogenated amorphous carbon. Hydrogenated DLC, produced via Chemical Vapor Deposition (CVD), has been developed and evaluated for its super-low friction performance in high vacuum. Typically, the hardness of CVD hydrogenated DLC is around 10 GPa, so a higher hardness hydrogenated ta-C exceeding 10 GPa is expected to provide low friction and high wear resistance in vacuum conditions. Moreover, such materials should be capable of operating at high temperatures, up to 200°C, in high vacuum environments.

In this study, ta-C coatings were prepared using the Filtered Cathodic Vacuum Arc (FCVA) method, with hydrogen ions from hydrogen (H_2) used for ion irradiation at various hydrogen atomic concentrations. Friction tests were then conducted on these ta-C coatings, which were sliding against bearing steel balls under both atmospheric pressure and high-vacuum conditions, varying from 23°C, 100°C, to 200°C. Following the friction tests, the role of hydrogen doping in ta-C for achieving low friction was analyzed and discussed.

2. Experimental methodology

2.1 Preparation of ta-C:H coating

The ta-C:H coatings were deposited using ion beam assisted-filtered arc deposition (IBA-FAD) machine equipped with hydrogen ion guns. The hybrid film deposition system enables the simultaneous deposition of carbon coatings (using T-shaped filtered cathodic vacuum arc deposition) onto an SUJ2 disk substrate, while hydrogen ion guns are employed during the process. A mirror finished SUJ2 disk (Φ22.5 mm,4 mm thick, surface roughness Ra below 0.01 μm, HRC60 or higher) was used as the substrate. Prior to deposition, the SUJ2 substrate was ultrasonically cleaned in benzene and acetone for 15 minutes each to remove the contaminants on the surface. The chamber was initially evacuated to approximately 0.004 Pa, with the working pressure fluctuating around 0.02 Pa during the deposition process.

The deposition was conducted in these processes: (1) pre-arc to clean the carbon target and pipe duct. (2) plasma etching using ion-gun, (3) Tantalum interlayer deposited by magnetron sputtering, and (4) ta-C and ta-C:H deposition. In details, argon ion plasma was carried out to etch the surface of SUJ2 disk to remove oxidants and contamination; argon gas was introduced at a flow rate of 16 sccm, with a discharge voltage set to 1.8kV. Argon gas (flow rate of 16 sccm) was ionized via glow discharge, and sputtering was performed for 15 minutes at a power output of 600W, resulting in a 500 nm thick tantalum interlayer. The deposition of ta-C:H coating was carried out with an arc discharge current of 50 A, along with a substrate bias voltage of -100 V and a substrate stage rotation speed of 10 rpm. Additionally, hydrogen gas (flow rate of 20 sccm) was introduced using the ion gun, with a discharge voltage of 1.8 kV.

2.2 Coating characteristics

The nano-hardness and elastic modulus of the coatings were evaluated using a Nano-Indenter (Elionix ENT-1100a, Japan). The film thickness, surface morphology, and wear track of the ta-C disk were analyzed using confocal laser scanning microscopy (Olympus OLS5100, Japan) and Field Emission Scanning Electron Microscopy (FESEM; Hitachi High-Technologies SU8230, Japan), equipped with energy-dispersive X-ray spectroscopy (EDS). The bonding states were analyzed through Raman spectroscopy (RENISHAW inVia Reflex, England) and X-ray photoelectron spectroscopy (XPS). Additionally, the atomic concentration of hydrogen in the ta-C film, across its depth, was measured using Rutherford Backscattering Spectroscopy (RBS) and Hydrogen Forward Scattering Spectroscopy (HFS).

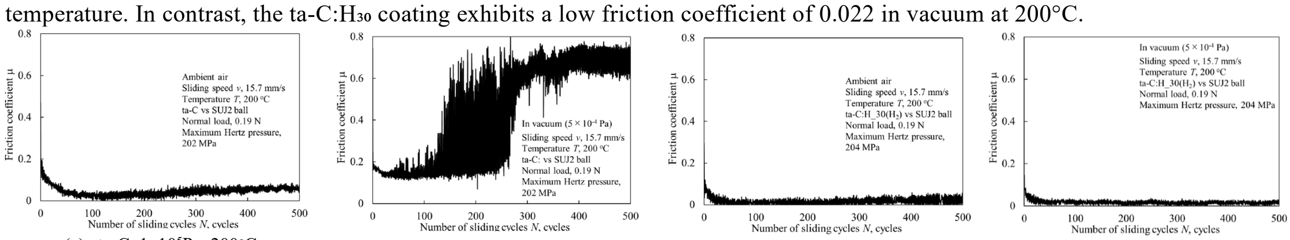
2.3 Friction test in high vacuum and high temperature

The tribological properties of the ta-C coatings under vacuum and high-temperature conditions were examined using a ball-on-disk friction tester. This equipment can achieve a high vacuum (5×10^{-4}Pa) in the chamber using a turbo molecular pump and can heat the specimen using an infrared heater (200°C), as shown in Fig. 1. An SUJ2 bearing steel ball (diameter = 8 mm) was used as the counterpart against the ta-C coating. Both the ball and the ta-C coated disk were cleaned in an ultrasonic bath with acetone for 15 minutes prior to testing. The coated disk was rotated at a sliding velocity of 15.7 mm/s for a total sliding time of 10 minutes, with a normal load of 0.19 N applied to the specimen using deadweight.

3. Results and discussions

3.1 Structural characteristics and tribological properties of ta-C:H coating

The hardness of the ta-C coatings decreased from 32.4 GPa to 20.1 GPa upon the introduction of 30 sccm hydrogen gas, resulting in a hydrogen content of 15.40% in the ta-C coating. However, the roughness (Rq) remained approximately 60 nm. Fig. 2 illustrates the variation in the friction coefficient of ta-C and ta-C:H_{30} coatings with the number of sliding cycles in ambient air and vacuum at 200°C. The results indicate that the vacuum environment significantly increases the friction coefficient of ta-C at this high temperature. In contrast, the ta-C:H_{30} coating exhibits a low friction coefficient of 0.022 in vacuum at 200°C.



(a) ta-C, 1×10^5Pa, 200°C (b) ta-C, 5×10^{-4}Pa, 200°C (c) ta-C:H_30(H_2), 1×10^5Pa, 200°C (d) ta-C:H_30(H_2), 5×10^{-4}Pa, 200°C

Fig. 2 Variation in the friction coefficient of ta-C and ta-C coatings with the number of sliding cycles in ambient air and vacuum at 200°C.

3.2 Structural characteristics and tribological properties of ta-C:H coating

After friction test, the worn part (wear track on ta-C coated disk and wear scar on SUJ2 ball) were observed using optical microscope. Raman spectroscopy and XPS analysis detected adhered graphite-like transfer films from ta-C:H coated surface, which was identified as the main factor contributing to low friction. Subsequently, the thickness of black transfer film was estimated from 3D imaged obtained using a laser optical microscope, as summarized in Fig.3. Fig.3 illustrates the relationship between average thickness of transferred film and average friction coefficient for ta-C:H_{10} and ta-C:H_{30} at various temperature in vacuum. It can be found that an increase in the thickness of the transferred film is necessary to reduce friction. For ta-C:H_{30}, an increase in film thickness with temperature corresponds to decreased friction. Conversely, for ta-C:H_{10}, a rise in temperature leads to an increase in film thickness, which also contributes to reduced friction. However, when the temperature exceeds 100°C, the growth of the transferred film thickness halts, resulting in increased friction for ta-C:H_{10}.

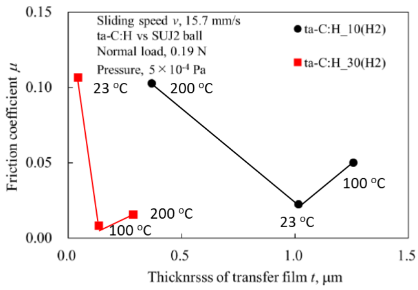


Fig.3 Relationship between the average thickness of the transferred film and the average friction coefficient for ta-C:H_{10} and ta-C:H_{30} at various temperatures in vacuum.

4. Conclusion

To achieve low friction and high wear resistance of Diamond-like Carbon (DLC) coatings in a vacuum at 5×10^{-4} Pa and high temperatures up to 200°C, hydrogen-doped ta-C coatings were deposited on SUJ2 steel plates using FCVA device. After conducting friction tests with the ta-C coated disks sliding against SUJ2 steel balls in vacuum and at high temperatures, it was found that the ta-C:H_{30} coatings are suitable for use in these conditions. However, in ambient air, ta-C:H_{10} could not achieve a friction coefficient lower than 0.1 due to hydrogen desorption occurring at 200°C, which resulted in increased friction. In summary, ta-C:H_{30} demonstrated a low friction coefficient of less than 0.1 across the temperature range of 23°C to 200°C, both in ambient air and vacuum conditions. Raman analysis revealed that the graphite-like transfer layer contributed to low friction, and a stable thick transfer layer maintained the low friction of ta-C:H_{30} even as the temperature increased to 200°C.

5. Reference

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