Effect of the substrate bias on tribological behavior of ta-C films at elevated temperatures
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ABSTRACT – In this study, the 1 µm of ta-C coating was deposited onto Inconel alloy disk by the FCVA technique with a different substrate bias of 0, 100 and 300 V. The tribological behaviors of ta-C coated disks sliding against Silicon Nitride (Si3N4) balls were examined under elevated temperature up to 600°C. The range of temperature was setting up until peel off observed. The experimental results showed that the friction coefficient was decreased from 0.2 to 0.08 with increasing temperature. At 600°C, the friction coefficient was dramatically increased over 4,000 cycles and then delaminated due to the graphitization. The wear rate showed a different behavior with a substrate bias. The wear rate of ta-C deposited of 0V was sharply increased at 100°C and then maintains a value in the range of 5×10⁻⁶ mm²/N·m. Meanwhile, it appears that increasing the temperature from 23 to 100°C increases wear rate at about 9×10⁻⁶ mm²/N·m. Further increase of that to 500°C leads to the decrease of wear rate. Raman spectroscopy and radius of curvature tech-nique were used to characterize the films structure and residual stress. Structural analysis revealed that thermal graphitization was occurred at 500°C. And the variation trend of the wear rate of ta-C films with increasing a temperature is similar to that of residual stress. Wear behavior of the ta-C fabricated with a different substrate bias was discussed in terms of residual stress, structural analysis.

1. INTRODUCTION

Diamond like carbon (DLC) films are consist of sp² and sp³ bonded carbon atoms with a high sp³ fraction, presenting a high hardness, high resistivity and low friction coefficient at room temperature. These properties make them suitable for many tribological applications such as wear resistance coatings for cutting tools and engine component [1]. However, those operations are performed at high temperatures with generating frictional heat during contact, these tribological properties of DLC films deteriorate rapidly [2,3].

Among the DLC series, Non-hydrogenated tetrahedral amorphous carbon (ta-C) is an ideal candidate for those application due to good thermal stability from high sp³ content, which contain more than 80 %. Various methods have been employed for depositing ta-C films, such as plasma-enhanced chemical vapor deposition (PEDVD), Filtered cathodic vacuum arc (FCVA) and sputtering deposition. Of these methods, FCVA deposition is a promising technique for the production of high quality ta-C coating [4]. And It has been reported by many researchers that by controlling the substrate bias, maximum the sp² and sp³ ratio was obtained [5].

The purpose of this study is to investigate the influence of the substrate bias potential from 0 to 300 V to understand the relationship which will in designing coatings between physical properties and tribological behavior under the high temperatures. And in this paper focused on ta-C coatings sliding against a Si3N4 by performing ball-on-disk experiments at elevated temperatures up to 600 °C to characterize the relationship between physical properties and tribological behavior.

2. EXPERIMENTAL DETAILS

2.1 Preparation of DLC film

The ta-C coatings investigated in this study were deposited on Inconel and Si substrates by using a FCVA system. It employs a 90° curved filter to remove the macro-particles. A rotated substrate holder was placed normal to the arc plasma beam at a distance of 15 cm from the exit of the FCVA source. Prior to the deposition, the substrate was thoroughly cleaned ultrasonically in alcohol and de-ionized water. The chamber was then evacuated to a base pressure of about 5 × 10⁻⁵ torr. Before deposition, Ar⁺ ion beam were used to remove the native oxide layer on the silicon and inconel surface. Cr layer was deposited on the substrate to promote adhesion of the ta-C film. Pure graphite (99.999%) was used as the cathode to obtain the carbon plasma. The films were deposited using a duct bias of 10 V at the filter. And a negative bias voltage was applied to the substrate via the substrate holder. The negative substrate bias was applied by using an asymmetric bipolar bias of 0, 100 and 300 V in the chamber.

2.2. Film characterization

The tribological behavior of ta-C films were confirmed by using a a high-temperature ball-on-disk tribo-meter to evaluate the friction between a ta-C coated disk and a Si3N4 ball in a high-temperature environment, as shown in Figure 1. These disks were heated to 23, 100, 200, 300, 400, 500 and 600°C,
respectively with the Infrared lamp and temperature maintained at a set value during the friction test. The Si₃N₄ ball (Φ=8 mm) was used as a counter-part against the rotating disk with a normal load of 1 N. All tests were conducted at a rotation rate of 200 rpm over 10,000 cycles and the track radius on the sample was 3 mm, unless the coating until peel off observed.

After tribo-test the microstructure of the wear track and wear scars on the film and ball measured by using Raman spectroscopy (Jasco NRS-1000), with 532 nm laser. The result of the wear tracks on the disks and wear scars on the Si₃N₄ ball being observed by optical microscopy. Quantitative wear rates were determined by calculating the volume of the wear tracks removed from the Non-contact surface profiler (ZYGO). The residual stresses of the films were measured using radius of curvature technique, which compares the curvature of the uncoated silicon substrate with the coated substrate. The radius of curvature of the substrates were measured using a Residual stress tester (J&L tech) and the compressive stress of the film is calculated using Stoney’s equation.

3. RESULTS AND DISCUSSION

Figure 2 shows the average friction coefficient of ta-C coating as a function of temperature from 23 to 500°C. A friction coefficient of ta-C deposited at 0V showed a tendency to increase up to 0.122 from room temperature to 300°C, then decrease slightly. Meanwhile, A friction coefficient of ta-C deposited at 100, 300 V showed a same tendency. Increasing the temperature from the 23 to 100°C results in an obvious increase of friction coefficient. However, as the temperature is further higher, the coefficient was gradually decreased.

Whereas in all samples at 600°C, the friction coefficient increases suddenly at 4000 or 5000 cycles, and which point, the test was stopped.

The wear rates of the ta-C coatings with a substrate bias of 0V are shown in Figure 3(a). At room temperature, the wear rate was the lowest at 0.49×10⁻⁶ mm³/N·m. The wear rate then increased to 4.45×10⁻⁶ mm³/N·m at 100°C, and the wear rate increased slightly to 6.57×10⁻⁶ mm³/N·m at 300°C. Then, at higher temperature up to 500°C, the wear rate maintained.

Figure 3(b) shows the wear rate of the ta-C coatings with a different substrate bias of 100, 300 V. Based on ta-C of 100 V, at room temperature, the wear rate was the lowest at 0.69×10⁻⁶ mm³/N·m. The wear rate was sharply increased to 9.22×10⁻⁶ mm³/N·m at 100°C, and then the wear rate decreased gradually to 1.92×10⁻⁶ mm³/N·m with increasing the temperature.
The Raman spectra of ta-C films deposited at different substrate bias of 0, -100, 300 V are shown in Figure 4(a). The Raman spectra show typical a characteristic ta-C film peak in this condition. The single broad peak can be deconvoluted into two peaks, one around 1350 (D-peak) and the other around 1580 (G-peak) [6]. Figure 4(b) shows that the ratio of intensities of the D-band to the G-band is also a function of the substrate bias. There was a decrease in the ID/IG ratio as substrate increased to substrate of -100 V. And then further increasing substrate bias of -300 V, I(D)/I(G) ratio slightly increased.

![Raman spectra](image1)

(a)

![Graph showing ID/IG ratio vs. substrate bias](image2)

(b)

Figure 4 (a) Raman spectra, (b) I(D)/I(G) ratio in the ta-C as a function of negative bias voltage

As suggested by Ferrari and Robertson, the measured ratios I(D)/I(G) can be used for qualitative interpretation of the amounts of sp³ and sp² bonds [7]. Fig. 4(b) shows that the lowest ratio ID/IG was obtained in ta-C coatings deposited at substrate bias of -100 V. This result suggests that these coatings fabricated at substrate bias of -100, 300 V have a higher ratio of sp³ ratio compared to the coatings obtained at value of 0 V. Confirmation of the assumption that at the lowest ratio I(D)/I(G) the deposited ta-C coating contains more sp³ bonds will be confirmed using additional studies by X-ray photoelectron spectroscopy.

Figure 5(a) show a surface image ta-C deposited with bias of 0V and conducted friction test at 500°C, and quite a few defects appear on the surface. To confirm structural change during the friction test, the coating surface was characterized by Raman spectroscopy. As shown in Figure 5(b), it supposed that the graphitization partly occurred on the surface [8].

![Surface image of ta-C](image3)

(a)

![Raman spectrum of defect](image4)

(b)

Figure 5 (a) Optical microscope image of the ta-C with bias of 0 V after friction test at 500°C and (b) Raman spectrum of defect on the surface
The tribological properties can be explained by considered the residual stress and structural change of ta-C film. As in previous studies, the friction coefficient is lower at the high temperature due to the formation of the transfer layer formed on the Si$_3$N$_4$ counterpart. And the wear rate at high temperatures was accelerated due to graphitization. However, in the case of ta-C fabricated at 100 V or higher, showed a different wear behavior. By applying the higher substrate bias, ta-C coating has a high sp$^3$ content due to the ion implantation effect [5]. However, high residual stress was strongly dependent with sp$^3$ content [9]. It seems to have had a dominant influence on the wear rate at 100 °C. As the temperature increases, the movement of carbon particles becomes active and then the residual stress decreases, and the wear rate decreases accordingly.

4. CONCLUSIONS

The study concludes that, ta-C coatings were deposited with a different substrate bias of 0, 100 and 300 V to investigate the tribological behavior under the high temperature condition. The experimental results showed that the friction coefficient was decreased from 0.2 to 0.08 with increasing temperature. At 600 °C, ta-C was delaminated from the substrate during the friction test due to the graphitization. The wear rate showed a different behavior with a substrate bias. The wear rate of ta-C deposited of 0V was increased at 100 °C and then maintains a value in the range of 5×10$^{-6}$ mm$^3$/N·m from the 200 to 500 °C. Meanwhile, It appears that increasing the temperature from 23 to 100 °C sharply increases wear rate at about 9×10$^{-6}$ mm$^3$/N·m. Further increase of that to 500 °C leads to the decrease of wear rate with a decrease of residual stress. These phenomena were summarized two kinds of reasons: (1) Graphitization and (2) Effect of residual stress on the ta-C coating. The ta-C of 0V having a low sp$^3$ content appeared to have a high wear rate at high temperatures due to the effect of graphitization. In the case of ta-C fabricated at 100 and 300 V, the high sp$^3$ content of the film serves to retard graphitization. At the same time, high residual stress strongly dependent with sp$^3$ content, it seems to have had a dominant influence on the wear rate.

REFERENCES