Jurnal Tribologi 23 (2019) 38-47

MYTRIBOS
Malaysian Tribology Society

Jurnal Tribologi
Website : jurnaltribologi.mytribos.org
e-issn: 2289-7232

Investigating running-in behavior to understand wear behavior of ta-C coating with filtered cathodic vacuum arc deposition

Woo-Young Lee [*], Takayuki Tokoroyama, Motoyuki Murashima, Noritsugu Umehara

Department of Mechanical Science and Engineering, Graduate School of Engineering,
Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8603, JAPAN.
[*]Corresponding author: Lwy13579@naver.com

KEYWORDS

ta-C coating
Defect
FCVA
Tribological behavior

ABSTRACT

In this study, to clarify running-in cycle and role of carbon transfer layer on wear, ta-C coatings were subjected to friction tests under 200 °C in air with a different sliding cycles. The steady state friction coefficient of 0.02 was obtained after finishing a running-in cycles approximately 2,000 cycles. During the steady state, wear rate of ta-C decreased with increasing number of cycles. Such the decrease of wear rate was explained by polished defects presented on surface and form carbon transfer layer on the counter-part material. The mechanism of those tribological properties was analyzed by Raman spectroscopy, scanning electron microscope observation and 3D measuring laser microscope.

1.0 INTRODUCTION

Diamond like-carbon (DLC) coating consists of sp^2 graphite structure and sp^3 diamond structure. These DLC coating was divided into the hydrogenated diamond-like carbon and non-hydrogenated amorphous carbon with a content of hydrogen and sp^2/sp^3 ratio (Bhowmick et al., 2015; Drescher et al., 1996; Enke et al., 1980). Among the DLC series, ta-C coating deposited by FCVA technique have a great attention in various fields such as infrared optical system, bio-medical application and auto-mobile component due to the high chemical inertness, mechanical and low frictional properties (Holmberg et al., 2000; Mabuchi et al., 2013; Robertson, 1986). Especially, ta-C coating was a good candidate as a protective coating to the tribological application demanding to the superior durability and low friction properties at high temperature condition (Robertson, 2008; Ronkainen & Holmberg, 2008; Sheeja et al., 2003).

In this regard, it is important micro structure of ta-C coating which is relationship between coating durability and performance of application. In additionally, the durability of ta-C coating

Received 20 March 2019; received in revised form 5 April 2019; accepted 22 July 2019.
To cite this article: Lee et al. (2019). Investigating running-in behavior to understand wear behavior of ta-C with filtered cathodic vacuum arc deposition. Jurnal Tribologi 23, pp.38-47.

depends on the surface roughness, defects like a pinhole and temperature. In previous research, many studies have been carried out to reveal the relationship between DLC coatings and wear properties. Y. Mabuchi et al. (Sheeja et al., 2001) reported the effect of the dropping out of droplets on ta-C coating fabricated by FCVA method under a lubricated condition. D. Drescher et al. (Sheeja et al., 2000) showed the morphology and structural properties of DLC films deposited by Laser arc arrangement. H. Takikawa et al. (Takikawa, 2006) analyzed the droplets generated during the deposition process using T-FAD system and suggest the minimization method of droplets. A. Dorner et al. (Waesche et al., 2009) deposited DLC coating with a different thickness and characterized the structural and durability of DLC coating as a function of coating thickness.

However, there are few investigations for effect morphology and defects in ta-C coating on wear and friction behavior. Therefore, the purpose of this study is to investigate the influence of defect and transfer layer on the friction and wear behavior which is able to help in understanding running-in cycles at high temperature condition. The main attention is paid to the mechanism of transfer layer and wear behaviour. And this paper focused on tribological behavior of ta-C coated disk against Si_3N_4 ball were carried out at high temperatures of 200 °C with different sliding cycles.

2.0 EXPERIMENTAL PROCEDURE

The ta-C coatings were fabricated on Inconel disk by using a FCVA techneque with a 45° curved filter employed to remove all unwanted macro-particles. A substrate holder was rotated at a constant speed for uniformity with the exit of FCVA source to substrate of 15 cm. Prior to the deposition, the substrate was thoroughly cleaned ultrasonically in alcohol and de-ionized water. The deposition base pressure was below 5×10^{-5} torr. Before deposition, ion beam with argon gas was employed to remove the oxide layer and impurities on surface of Inconel disk composed with nickel (68 %), chrome (17 %) and iron (8%). Inconel disk is quite suitable for this study requiring high strength and good corrosion resistance in high temperature due to investigating tribological behavior of top surface of ta-C coating. A chromium layer was deposited to improve the adhesion between the substrate and ta-C coatings. ta-C coating was deposited using a negative substrate bias voltage of 100 V.

The tribological performance of ta-C coatings was confirmed by using a high-temperature ball-on-disk type tribo-tester to evaluate the friction. These specimens were preheated 200°C, in ambient air via an Infrared lamp and temperature was maintained at the set value during the friction test. A silicon nitride (Si_3N_4) ball (Φ=8 mm) was used as a counter-part material with the wear track radius of 3 mm. For each test, a load of 1 N was applied on a Si_3N_4 ball rotating at a sliding speed of 200 rpm until completion of 2,000, 4,000, 8,000 and 12,000 cycles, respectively. The microstructure of the wear tracks on the coating and wear scars on Si_3N_4 ball were measured by using Raman spectroscopy (Jasco, NRS-1000), with 532 nm laser and examined by Scanning electron microscope (SEM) and optical microscopy. To calculate the cross-sectional of wear tracks measured by Non-contact surface profiler (ZYGO, Newview6200), an average of four different measurements was used. The thickness and surface roughness of transfer layer was confirmed by 3D measuring laser microscope (Olympus, LEXT OLS5000)

3.0 RESULTS

Figure 1 showed friction coefficient of the ta-C coatings fabricated with a substrate bias voltage of -100 V up to sliding cycles from 2,000 to 12,000 under temperature of 200 °C. In this experiment, ta-C coating exhibited a running-in cycles and then the steady state (μ_S=0.02) was reached after 2,000 cycles. The other result with a different number of cycles of 4,000, 8,000 and 12,000 showed similar behavior that the running-in cycles was maintained up to 2,000 cycles and steady state was shown.

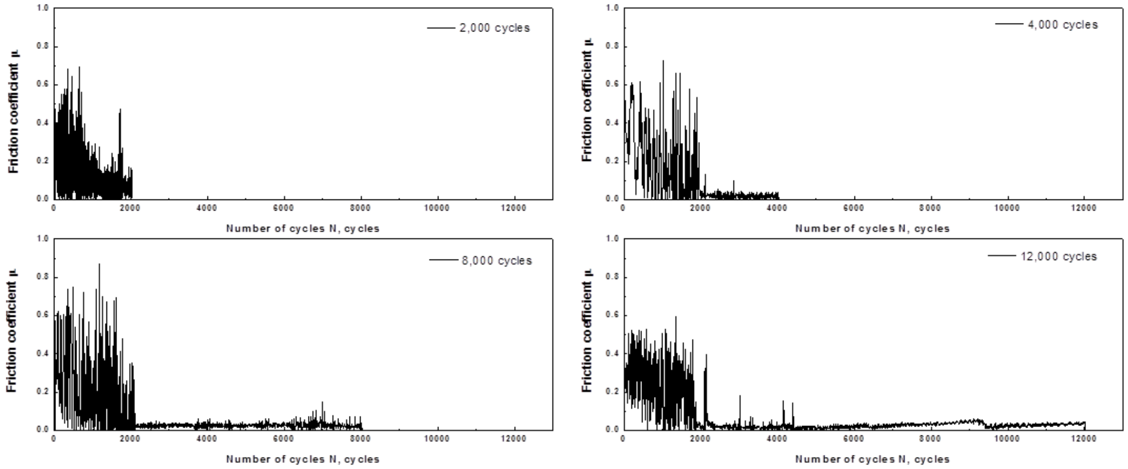
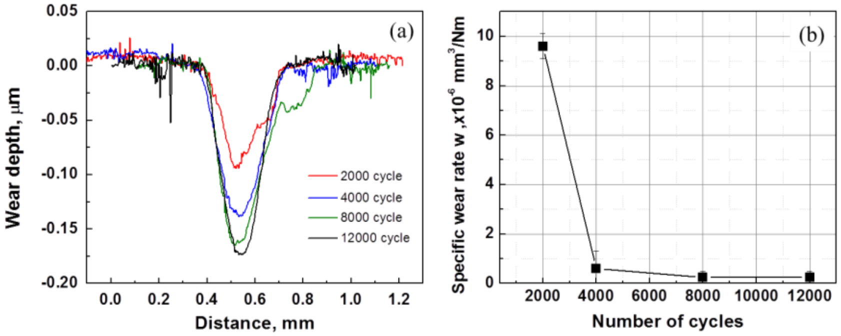


Figure 1: Friction coefficient of ta-C against the Si_3N_4 ball at 200 ℃. Tests were run for 2,000, 4,000, 8,000 and 12,000 cycles, respectively.

The combined two-dimensional cross section image of wear track of ta-C coating for 2,000, 4,000, 8,000 and 12,000 cycles are shown in Figure 2(a). At 2,000 cycles, ta-C coating surface was considerable damaged with a fluctuation of friction coefficient. However, the depth of wear track became slightly deeper with an increase of sliding cycles. A specific wear rates can be calculated using the cross-sectional wear area with length of wear track and applied load. And the wear rate of the 4,000, 8,000 and 12,000 cycles was calculated except to the wear volume occurred in 2,000 cycles to confirm the effect of the transfer layer on wear behavior. The ta-C coating exhibited a higher wear rate of 9.6x10^{-6} mm^3/Nm at 2,000 cycles. However, wear rate started to be sharply decreased to 6.0x10^{-7} mm^3/Nm from 2,000 to 4,000 cycles and then reached to the 2.5x10^{-7} mm^3/Nm from 2,000 to 12,000 cycles as shown in Figure 2(b).



(a) (b)

Figure 2: (a) Combined cross-sectional image of wear track, (b) specific wear rate of ta-C coating as a function of number of cycles.

Figure 3(a) showed surface morphology of ta-C coating measured by SEM micrograph. Through the SEM image, various shape and sizes of pores and nodular defects were visible. Some of nodular defects called as droplets looked like that macro-particles were embedded in ta-C matrix. The sized of nodular defects varied around a value of 0.7-1.0 μm. There were probably also some pores formed when the solidified droplets break off after the deposition process has been completed. Some pores with various diameters of 0.05-10 μm were present. To confirm the effect of defects on tribological behavior, surface morphology at designated area was compared with increasing different sliding cycles. After 1,000 cycles, change of surface morphology in wear track of ta-C coating was observed. It is notice that top surface of nodular defects were polished but were remained inside of wear track as presented in Figure 3(b). And then nodular defects were almost grinded off. Pores and scratch grooves damaged by abrasive wear were confirmed along with the sliding direction displayed in Figure 3(c). Especially, these changes of morphology in nodular defect could be seen more clearly in enlarged SEM image as shown in Figure 4. The nodular defect was survived at 1,000 cycles and then grinded after finishing running-in cycles.

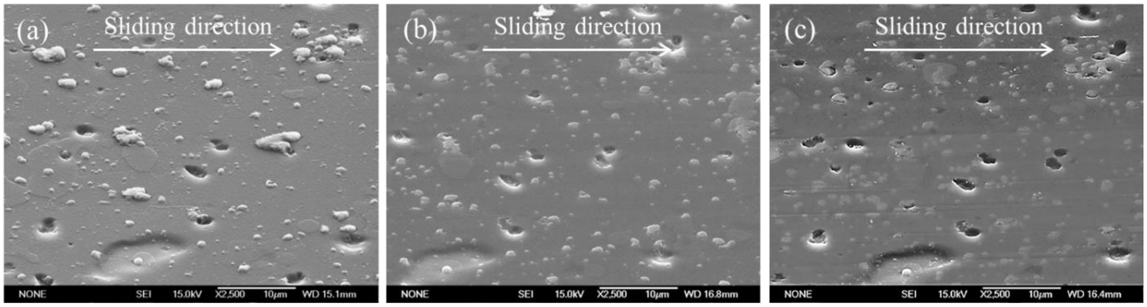


Figure 3: SEM image of wear track at different sliding cycles at (a) 0 cycles, (b) 1,000 cycles and (c) 4,000 cycles.

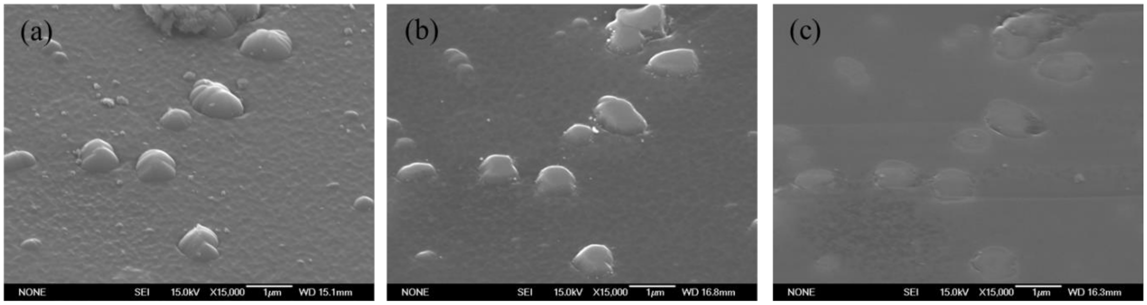


Figure 4: Enlarged SEM image of wear track at a different sliding cycles at (a) 0 cycles, (b) 1,000 cycles and (c) 4,000 cycles.

After the friction test, there were clearly noticeable wear scar on surface of the Si_3N_4 ball. Figure 5(a) shows the optical image of wear scar on the Si_3N_4 ball in the contact area at 2,000 cycles. It is obviously seen that wear scars, partially covered by multi-color layer with a rough surface, formed on the Si_3N_4 ball. At the same time, there was an accumulation of carbon debris in front of wear scar. At 4,000 cycles, these were noticeable tribo-layer formation with a partial buckling and an amount of wear debris was more accumulated. And further increase of sliding cycles up to 12,000 cycles, tribo-layer could be seen on wear scar without bucking compared to that at 4,000 cycles. The variation of diameter of wear scars was 422, 386 and 388 μm at 2,000, 4,000 and 12,000 cycles, respectively. No obvious change was observed for the diameters of wear scars from 4,000 cycles and 12,000 cycles.

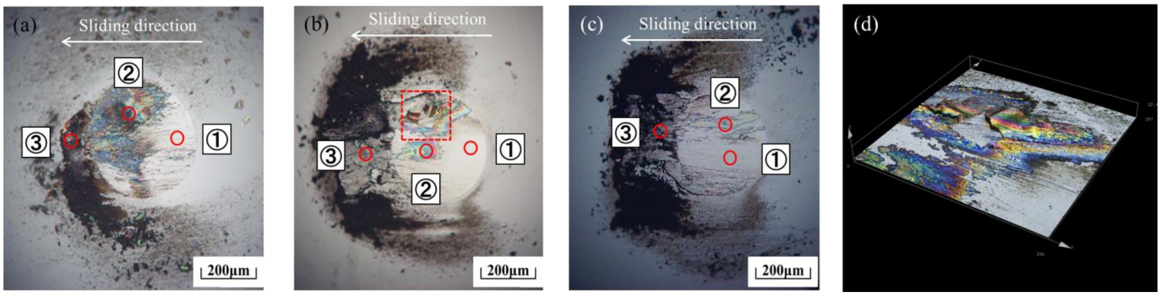


Figure 5: Wear scar on the ball at different sliding cycles at (a) 2,000 cycles, (b) 4,000 cycles and (c) 12,000 cycles. The small area indicated in red square in Fig. 5 (b) is enlarged on the right.

In order to confirm the carbonaceous tribo-layer on wear scar and explain the tribological behaviour with a formation of transfer layer, Raman analysis was conducted. Figure 6 show the difference of Raman spectrum for ta-C coating, Si_3N_4 ball and graphitized transfer layer on wear scar of Si_3N_4 ball against ta-C coating with sliding cycle of 4,000 cycles. In the case of graphitized transfer layer on Si_3N_4 ball, the D peaks around 1350 cm^{-1} and G peaks around 1580 cm^{-1} was separated. This means that the graphitized transfer films are formed wear scar.

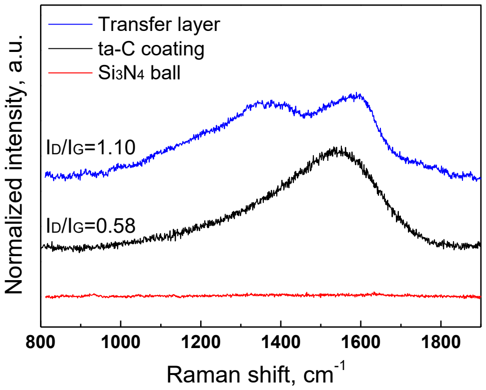


Figure 6: Raman spectrum of Silicon nitride, as-deposited ta-C coating and graphitized transfer layer on the wear scar after testing up to 4,000 cycles.

The Raman spectra of carbon debris accumulated in front of sliding direction, multi-color layer presented on the wear scar and polished surface was investigated, as depicted in Figure 7. The Raman spectra in Figure 7(a) showed a characteristic of graphitization in the carbon debris and multi-color layer. The appearance of D peak at 1350 cm^{-1} with a low intensity in the Raman spectra on the carbon debris and multi-color layer indicated an increase of sp^2 bonds with a change in the bonding structure of the surface of ta-C coating and small size of cluster. However, polished surface beside the transfer layer only indicated the Si_3N_4 peak (Deng et al., 2014). Further increase of sliding cycles until 4,000 cycles, higher intensity of Raman spectrum was shown compared to that up to 2,000 cycles. And it is important to notice that carbonaceous transfer layer could be seen not only at multi-color layer but also polished area at 12,000 cycles. From this observation, it was obviously seen that wear scars was partially covered by carbonaceous transfer films in initial term. During steady state wear scar was overall covered by transfer layer. The transfer layer became a smooth surface and approximately maintains a thickness of around 200 µm as shown in Figure 8. The surface roughness of transfer layer on Si_3N_4 ball showed a decreasing trend in a range from 74 to 38 nm.

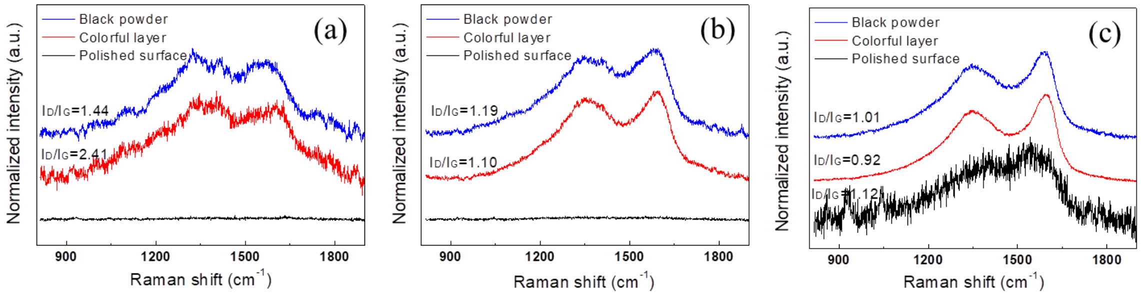


Figure 7: Raman spectrum of carbon debris, colorful layer and polished surface on the wear scar after testing up to (a) 2,000 cycles, (b) 4,000 cycles and (c) 12,000 cycles.

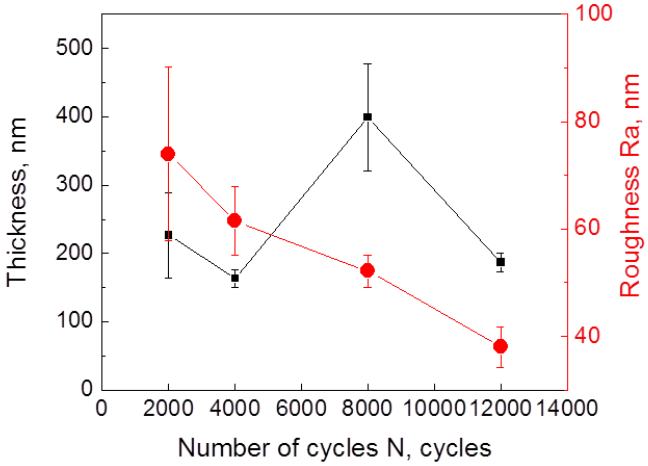


Figure 8: Comparison between thickness and roughness of transfer layer as a function of sliding cycles.

4.0 DISCUSSION

To reveal the factor the running-in cycles and understand mechanism of transfer layer formation on the counterpart material, morphology, friction and wear behavior of ta-C coating against Si_3N_4 ball was conducted at 200 °C as a function of different sliding cycles. Noticeable tribological behaviour was that at the beginning of the sliding, running-in cycles and severe wear of ta-C coating was shown up to 2,000 cycles. During the steady state from 2,000 to 12,000 cycles, friction coefficient and wear rate was dramatically decreased.

On the basis of SEM microscopy (Figure 3(a) and Figure 4(a)), a lot of defects was observed on the ta-C coating surface. These nodular defects were survived at 1,000 cycles. Nodular defect on the ta-C coating caused running-in cycles and higher wear rate until the 2,000 cycles. And abrasive particles spalled off nodular defects might result in the severe wear at sliding interface shown in Figure 3(c). Rough surface of tribo-layer on counter-part material was shown in Figure 5(a) and could be estimated from low intensity of Raman spectra illustrated in Figure 7(a). Transfer layer formed on wear scar of Si_3N_4 ball was peeling off by the nodular defects in the middle of running-in cycles. The nodular defect on ta-C coating surface is supposed to be the main reason for the running-in cycles with disturbing to form the tribo-layer on the counter-part material.

From the optical image of wear scar (Figure 5(b)) and Raman spectrum (Figure 7(b)), these are clearly noticeable formation of transfer layer corresponding to the low friction coefficient (0.02 shown in Figure 1) and lower specific wear rate after 2,000 cycles. Due to the presence of transfer layer formed on the wear scar of the ball, it is enough to passivate dangling bond generated at high temperature condition with a decreasing a shear strength and a protection of both ta-C coating and the Si_3N_4 ball surface.

The graphitized transfer layer on the Si_3N_4 ball is an important role of wear behaviour. To transfer the carbonaceous tribo-layer on the Si_3N_4 ball from ta-C coating, graphitized carbon debris was accumulated in front of sliding direction of Si_3N_4 ball. This accumulated debris protects the ball surface and be a source of the carbonaceous tribo-layer on Si_3N_4 ball during the sliding. In an initial term up to 2,000 cycles, this transfer layer was composed of small size of cluster with a rough surface, despite the presence of transfer layer. After that, Raman spectra of polished surface on ball at 2,000 cycles and 12,000 cycles were distinctly different. This means that thickness and structure of transfer layer was stably formed with thicker tribo-film and covering the whole of wear scar on the Si_3N_4 ball.

To account for friction and wear properties and a process of formations of carbonaceous transfer film by consideration of those defects, wear mechanism was proposed as schematic image expressed in Figure 9: The nodular defect become abrasive particle, causing the running-in cycles and severe wear (Figure 9(a) and (b)). During the steady state, growth of transfer layer protect the ta-C coating surface and Si_3N_4 ball with maintaining the thickness over 200 μm, covering the area and good adhesion properties as illustrate in Figure 9(c).

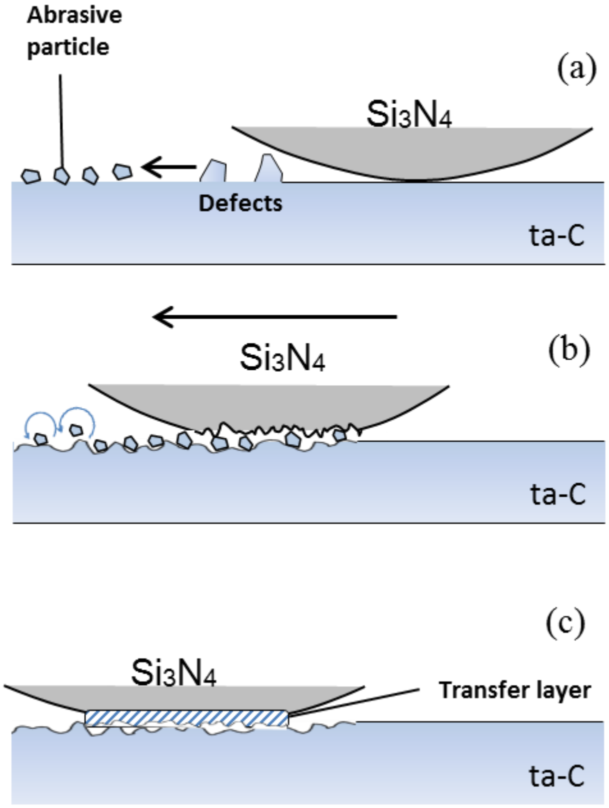


Figure 9: Schematic of wear mechanism at different sliding cycles; (a) as-deposited, (b) running-in cycles up to 2,000 cycles, (c) steady state from 2,000 to 12,000 cycles

5.0 CONCLUSION

The study presented the tribological behaviors of ta-C coating versus Si_3N_4 ball at high temperature as a function of sliding cycles. A conclusion is that tribological behaviors at high temperature are affected by morphology of ta-C coating surface and formation transfer layer. As a result, running-in cycles shown up to 2,000 cycles was finished with a removal of nodular defects and then specific wear rate was sharply decreased during the steady state region. Low friction coefficient of 0.02 corresponding to lower wear rates of 2.5x10^{-7} mm^3/Nm were observed at 12,000 cycles. Such low friction coefficient and decrease of wear rate was attributed by the formation of transfer layer on the wear scar of the Si_3N_4 ball. This graphitized transfer layer around thickness of 200 nm can significantly protect the ta-C coating surface with covering whole area of Si_3N_4 surface. The results demonstrate that transfer layer formation influence tribological behavior decreasing the interaction between ta-C coating and wear scar on the Si_3N_4 ball.

ACKNOWLEDGEMENT

This study was supported financially by the Fundamental Research Program (KIMS Academy Lab., PNK5960) of the Korean Institute of Materials Science and Otsuka Toshimi Scholarship Foundation.

REFERENCES

Bhowmick, S., Banerji, A., Khan, M. Z. U., Lukitsch, M. J., & Alpas, A. T. (2015). High temperature tribological behavior of tetrahedral amorphous carbon (ta-C) and fluorinated ta-C coatings against aluminum alloys. Surface and Coatings Technology, 284, 14-25.

Deng, X., Kousaka, H., Tokoroyama, T., & Umehara, N. (2014). Tribological behavior of tetrahedral amorphous carbon (ta-C) coatings at elevated temperatures. Tribology international, 75, 98-103.

Drescher, D., Scheibe, H. J., Mensch, A., Alers, P., & Dyer, C. (1996). Morphology and structural characterization of plasma-assisted prepared carbon films. Diamond and Related Materials, 5(9), 968–972

Enke, K., Dimigen, H., & Hübsch, H. (1980). Frictional properties of diamondlike carbon layers. Applied Physics Letters, 36(4), 291–292.

Holmberg, K., Ronkainen, H., & Matthews, A. (2000). Tribology of thin coatings. Ceramics International, 26(7), 787–795.

Mabuchi, Y., Higuchi, T., Inagaki, Y., Kousaka, H., & Umehara, N. (2013). Wear analysis of hydrogen-free diamond-like carbon coatings under a lubricated condition. Wear, 298–299(1), 48–56.

Robertson, J. (1986). Amorphous carbon. Advances in Physics, 35(4), 317–374.

Robertson, J. (2008). Comparison of diamond-like carbon to diamond for applications. Physica Status Solidi (A) Applications and Materials Science, 205(9), 2233–2244.

Ronkainen, H., & Holmberg, K. (2008). Environmental and thermal effects on the tribological performance of DLC coatings. Tribology of Diamond-Like Carbon Films: Fundamentals and Applications, (Dlc), 155–200.

Sheeja, D., Tay, B. K., Krishnan, S. M., & Nung, L. N. (2003). Tribological characterization of diamond-like carbon (DLC) coatings sliding against DLC coatings. Diamond and Related Materials, 12(8), 1389–1395.

Sheeja, D., Tay, B. K., Lau, S. P., & Shi, X. (2001). Tribological properties and adhesive strength of DLC coatings prepared under different substrate bias voltages. Wear, 249, 433–439.

Sheeja, D., Tay, B. K., Lau, S. P., Shi, X., & Ding, X. (2000). Structural and tribological characterization of multilayer ta-C films prepared by filtered cathodic vacuum arc with substrate pulse biasing. Surface and Coatings Technology, 132(2–3), 228–232.

Takikawa, H. (2006). Review of cathodic arc deposition for preparing droplet-free thin films. Proceedings - International Symposium on Discharges and Electrical Insulation in Vacuum, ISDEIV, 2(4), 525–530.

Waesche, R., Hartelt, M., & Weihnacht, V. (2009). Influence of counterbody material on wear of ta-C coatings under fretting conditions at elevated temperatures. Wear, 267(12), 2208–2215.