# Tribological properties of multilayer tetrahedral amorphous carbon coatings deposited by filtered cathodic vacuum arc deposition

Young-Jun JANG<sup>1,\*</sup>, Jae-Il KIM<sup>2</sup>, WooYoung LEE<sup>2</sup>, Jongkuk KIM<sup>1,\*</sup>

**Abstract:** Tetrahedral amorphous carbon (ta-C) has emerged as an excellent coating material for improving the reliability of application components under high normal loads. Herein, we present the results of our investigations regarding the mechanical and tribological properties of a 2- $\mu$ m-thick multilayer ta-C coating on high-speed steel substrates. Multilayers composed of alternating soft and hard layers are fabricated using filtered a cathodic vacuum arc with alternating substrate bias voltages (0 and 100 V or 0 and 150 V). The thickness ratio is discovered to be 1:3 for the sp²-rich and sp³-rich layers. The results show that the hardness and elastic modulus of the multilayer ta-C coatings increase with the sp³ content of the hard layer. The hardness reached approximately 37 GPa, whereas an improved toughness and a higher adhesion strength (> 29 N) are obtained. The friction performance ( $\mu$  = 0.07) of the multilayer coating is similar to that of the single layer ta-C thick coating, but the wear rate (0.13 × 10<sup>-6</sup> mm³/(N·m)) improved under a high load of 30 N. We further demonstrate the importance of the multilayer structure in suppressing crack propagation and increasing the resistance to plastic deformation (H3/E2) ratio.

**Keywords:** tetrahedral amorphous carbon (ta-C); filtered cathodic vacuum arc deposition; multilayer coatings; alternating substrate bias voltage; wear resistance; plastic deformation resistance

## 1 Introduction

To improve the reliability of application components under high normal loads, various thick protective coatings are applied. Tetrahedral amorphous carbon (ta-C) has emerged as an excellent coating material for mechanical machinery and automobile components, as it provides enhanced durability owing to its numerous sp³ bonds.

Although ta-C coatings are known to provide low friction and high wear resistance owing to their high hardness, thick ta-C coatings ( $\geq 1~\mu m$ ) have high internal stresses, which often result in weak adhesion

between the coating and substrate. These issues have been successfully addressed by controlling both the internal stresses of the coating and the surface temperature [1, 2].

Several studies have described the use of different coating methods such as single layer, multilayer, metal doping, and structural combination. They have primarily focused on hydrogenated amorphous carbon, amorphous carbon, and thin ta-C coatings ( $\leq 1~\mu m$ ) [3–11] because it is extremely difficult to control internal stresses arising from thicker coatings ( $\geq 1~\mu m$ ), particularly when the physical vapor deposition coating process involves a solid target. The use of thin

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coatings severely limits the mechanical properties of downsized mechanical components when they are exposed to extreme environments. Therefore, thick ta-C coatings ( $\geq 1~\mu m$ ) must be investigated to produce reliable mechanical components for industrial applications.

Among the various mechanical properties that determine the reliability of mechanical components, wear resistance is one of the most important factors requiring attention. It can affect several key physical properties such as roughness, surface quality, vibration of machineries, and component lifetime. To improve the wear resistance of coatings, the hardness, fracture toughness, and resistance to plastic deformation ( $H^3/E^2$ ) values should be optimized. Without using dopants, single layered ta-C coatings cannot satisfy these requirements. It has been reported that multilayer coatings can be used to optimize these properties [12–17].

Although dopants significantly improve the wear resistance of mechanical components, an expensive coating system equipped with multiple sources is required to perform doping during coating. Therefore, a thick coating that involves the use of a single material such as carbon can be a promising method for improving the quality of mechanical components.

The primary objective of the present study is to increase the wear resistance of engineering metals by ta-C coatings using multilayered structures as ta-C/ta-C combinations with different deposition parameters and thickness ratios. First, these coatings were deposited on high-speed steel (HSS) substrates using an alternating substrate bias voltage of 0:100 and 0:150 V. For a better surface finish after the coating, filtered cathodic vacuum arc (FCVA) deposition was employed. Thick ta-C coatings with a single deposition parameter without an alternating substrate bias voltage were prepared for comparison. The structural, mechanical, and tribological properties of these coatings were investigated under heavy normal loads to demonstrate their improved wear resistance.

# 2 Experimental

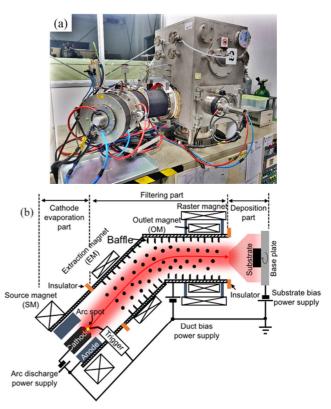
## 2.1 ta-C coating preparation

The ta-C coatings investigated in this study were deposited onto HSS substrates using an FCVA

deposition system, as shown in Fig. 1. The system comprised a vacuum arc source with a 45°-bent filter attached to the coating chamber. A carbon target of diameter 50 mm and purity of 99.99% was used. The magnetic coil current was fixed to 5 A.

Prior to the deposition, the HSS substrates were rinsed with alcohol and deionized water and then placed into a holder positioned perpendicular to the carrier to allow rotation during the deposition. After pumping out the chamber to a pressure below  $5 \times 10^{-3}$  Pa, the HSS substrates were etched for 20 min in Ar plasma at  $2.0 \times 10^{-1}$  Pa. A titanium interlayer was deposited to promote adhesion by unbalanced magnetron sputtering. Carbon arc deposition was performed using a duct bias of +15 V at the filter and a 60 A arc current with a pressure below  $1 \times 10^{-3}$  Pa. Detailed experimental methods have been described in previous studies [18, 19].

During the deposition of the thick multilayer ta-C coating, alternating negative substrate bias voltages ( $V_b$ ) of 0:100 and 0:150 V were applied using an asymmetric bipolar pulsed power supply (20 kHz, 90% duty cycle). A high  $V_b$  resulted in ta-C coatings with high sp<sup>3</sup>



**Fig. 1** (a) 45°-bent FCVA deposition system and (b) schematic illustration of carbon arc source.

fractions, whereas a low  $V_b$  resulted in coatings with high sp² fractions. The multilayered ta-C coatings comprised alternating layers of sp²-rich (soft) and sp³-rich (hard) coatings. The soft-to-hard ta-C coating thickness ratio was fixed to 1:3, whereas  $V_b$  was alternately varied between 0:100 and 0:150 V. In the case of a single layer ta-C coating, three different  $V_b$  values were used at 0, 100, and 150 V. The soft-to-hard ta-C coating thickness ratio was set to 1:3 to improve the residual stress and fracture characteristics of hard ta-C. If the thickness of the soft layer is thicker than that of the hard layer, then the hardness of the coating will deteriorate. Therefore, a thickness ratio of 1:3 was set to reflect the hard ta-C characteristics. The experimental conditions are summarized in Table 1.

## 2.2 Characterization of ta-C coatings

The thicknesses of the different ta-C coatings were measured via field emission scanning electron microscopy (FE-SEM, Tescan, Republic of Korea). Their hardness and elastic modulus were measured using a nano-indenter (Elionix Co., Ltd., Japan) with a Berkovich tip with confirmation of pop-in events. The hardness and elastic modulus of the coating film were measured by excluding the effect of the substrate by creating an indentation depth that was 1/10 of the coating thickness. The final load and the maximum indentation depth were 274 mN and 1.0  $\mu$ m for the composite hardness and elastic modulus, respectively. The Poisson ratio of the ta-C coatings was assumed to be 0.3 [20]. The maximum Hertzian contact pressure

**Table 1** Experimental conditions and properties of ta-C coatings.

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Sample No.	#1	#2	#3	#4	#5
Total coating thickness	2 μm	2 μm	2 μm	2 μm	2 μm
Structure	Single layer	Single layer	Single layer	Multilayer	Multilayer
Thickness ratio (soft layer: hard layer)	_	_	_	1:3	1:3
Cycle	_	_	_	5	5
Substrate bias	0 V	100 V	150 V	$0 \leftrightarrow 100 \text{ V}$	$0 \leftrightarrow 150 \text{ V}$
Interlayer	Ti	Ti	Ti	Ti	Ti
Substrate	HSS	HSS	HSS	HSS	HSS

 $(p_{\rm m})$  was calculated using the following equation:  $p_{\rm m} = 4E^*a/3\pi R$ , where  $E^*$  is the reduced elastic modulus, a is the real contact radius, and R is the equivalent curvature radius.

The physical structure of the coatings was analyzed via a Raman spectroscope (Horiba Co., Ltd., Republic of Korea) using Ar laser. An excitation wavelength of 514 nm was used in backscattering geometry mode at 23 °C and 40% relative humidity. The incident power was set at 2 mW, and the scanning range was 800–2,000 cm<sup>-1</sup>.

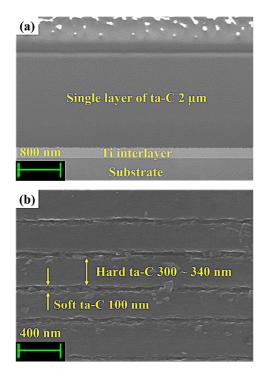
The adhesion properties of the ta-C coatings were evaluated using a scratch tester (J&L Tech, Republic of Korea). The friction and wear properties were determined using a ball-on-disc type tribometer. The flat-coated disks were mounted on a steel holder, which was fixed to a rotation holder. An  $Al_2O_3$  ball (diameter = 6 mm, Vickers hardness = 14 GPa [21]) was positioned against a disc, which was located 4 mm eccentrically from the center of the coated disc. A normal load of 30 N was applied to push the ball downward while the disc was rotated at 100 rpm for 10,000 cycles. The total sliding distance was 188 m, and tribology tests were performed thrice to verify the repeatability.

After performing those tests, the wear rate of the film was calculated based on the profile of the wear track measured using a confocal microscope (Olympus, Japan). The wear volume of each wear track was obtained as the average of three to five measurements in randomly selected areas. The wear rate (K) of the coating was calculated from the wear volume using the following equation: K = V/SF, where V is the wear volume in mm<sup>3</sup>, S is the total sliding distance in M, and M is the normal load in M. FE-SEM equipped with energy dispersive X-ray spectroscopy (EDS) was used to characterize the wear track after the tribology test.

### 3 Results and discussion

## 3.1 Thickness and structure of ta-C coating

Figure 2 shows the cross-sectional FE-SEM images of the single layer and multilayer ta-C coatings on HSS substrates. The thicknesses of the soft and hard layers of the multilayer ta-C are shown in Fig. 2(b). The soft

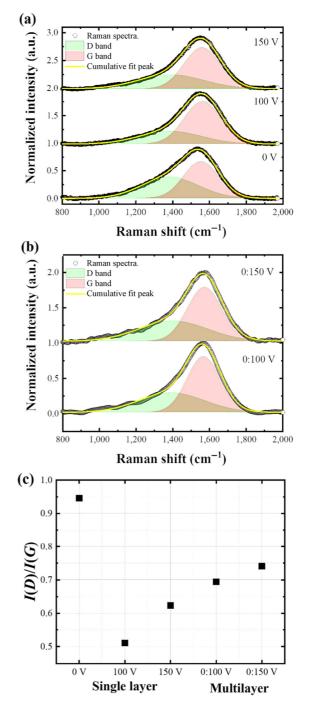


**Fig. 2** FE-SEM cross-sectional images of (a) single layer ta-C coating deposited with 0 V substrate bias and (b) multilayer ta-C coating deposited using alternating substrate bias (0:100 V).

layer (100-nm thick) was deposited at 0 V, whereas the hard layer (300–340 nm thick) was deposited at  $100 \, \text{V}$  of  $V_b$ . After a single cycle involving the deposition of one hard and one soft layer, the thickness of the ta-C coating was ~400 nm. Five such cycles were completed for each batch, resulting in a 2.0- $\mu$ m-thick coating, consistent with the experimental design. Moreover, thick and well-adhered single layer ta-C coatings were successfully produced via FCVA deposition, indicating reduced stresses compared with coatings deposited using other methods.

The results obtained from the Raman spectroscopy analysis of the ta-C coatings are shown in Fig. 3. The Raman spectra comprised a broad peak between 1,000 and 1,800 cm<sup>-1</sup>, which resulted from the superposition of a D-peak and a normal G-peak at approximately 1,350 and 1,580 cm<sup>-1</sup>, respectively.

In the case of ta-C coatings, the G-peak was assigned to the stretching of  $\mathrm{sp^2}$  bonds in both rings and chains, whereas the D-peak was assigned to the breathing modes of  $\mathrm{sp^2}$  bonds in rings. The Raman graphs were fitted using Gaussian functions, which were then used to determine the I(D)/I(G) ratio, as shown in Fig. 3(c). It is evident from Fig. 3(c) that the value of I(D)/I(G) for



**Fig. 3** Raman spectra obtained from (a) single layer and (b) multilayer of ta-C coatings. (c) I(D)/I(G) ratio as a function of bias voltage.

a single layer of ta-C coating decreased when  $V_b$  increased from 0 to 100 V. However, beyond 100 V, it increased with  $V_b$  until 150 V. The I(D)/I(G) ratio of the single layer ta-C coating deposited at 100 V had a relatively small value of 0.51. Because deposition with a  $V_b$  of 100 V resulted in layers with high sp<sup>3</sup>

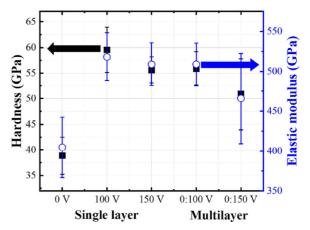
content, these layers tended to exhibit higher internal stresses [22].

For multilayer coatings deposited with alternating  $V_b$  of 0:100 and 0:150 V, the I(D)/I(G) ratios were 0.69 and 0.74, respectively. This implies that although the thickness of the hard layer increased, the soft layer (0 V) with increasing sp² content resulted in the release of internal stresses. This low internal stress can affect the mechanical properties such as hardness, elastic modulus, and toughness of coating.

## 3.2 Mechanical properties of ta-C coatings

Figure 4 shows the variation in nanohardness in the multilayered ta-C coating. The multilayer shows a mixed value of hardness of each single layer.

As shown in Fig. 5, the composite hardness and



**Fig. 4** Mechanical properties of single layer and multilayer ta-C coatings.

elastic modulus were measured by including the effect of the substrate. For the single layer coatings, it was

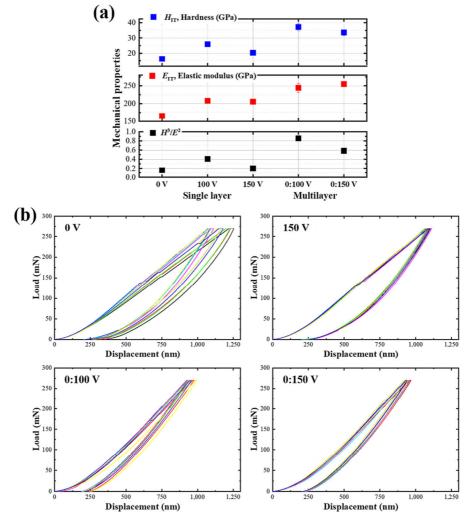


Fig. 5 (a) Composite mechanical properties and (b) force-displacement curves of single layer and multilayer ta-C coatings with high indentation load.

observed that when  $V_{\rm b}$  increased from 0 to 100 V, the hardness increased from 16 to 25 GPa. However, as  $V_{\rm b}$  increased to 150 V, the hardness decreased to 20 GPa. In addition, the behavior of the elastic modulus and the  $H^3/E^2$  values matched well with the variation in nanohardness corresponding to different  $V_{\rm b}$  values. These results were consistent with those obtained from the Raman analysis of the coatings.

The  $H^3/E^2$  ratio is physically used as a plastic index and resistance to plastic deformation, and it has been reported to be correlated with fracture because the yield pressure of ball contact during elastic/plastic deformation is a function of  $H^3/E^2$  [22, 23]. In a multilayer of relatively ductile (soft) ta-C and brittle (hard) ta-C, local deformation occurs via ductile ta-C, thereby reducing the bending stress. When a normal load is applied, the dislocation line energy is propagated [24]. In a multilayered structure, resistance of dislocation flow across other layers occurs, and each layer serves as an element that interferes with dislocation motion. In the case of multilayer coatings with a soft-to-hard layer thickness ratio of 1:3 at 0:100 and 0:150 V, the mechanical properties were better than those of the single layer coatings. The elastic modulus and  $H^3/E^2$ values were significantly higher than those of the single layer coatings. These results indicate that the soft-hard multilayered structure of the ta-C coating effectively prevented the deformation of the coated layers. Furthermore, a high value of  $H^3/E^2$  was obtained in the case of the 0:100 V ta-C coating. As such, deformations such as crack propagation between the interlayer of soft and hard ta-C, which served as an obstacle, were prevented. This can result in an increase in the nanohardness when an alternating  $V_{\rm b}$ is employed.

Figure 5(b) shows the representative force-displacement curves of the indentations rendered on the ta-C coatings. These curves indicate the elastic and plastic deformations that occur during indentation. All the coatings showed typical curves, characteristic of hard elastoplastic materials; however, the multilayer coatings exhibited higher elasticity than the single layer coatings. Moreover, in the case of single layer coatings, cracks appeared during loading at indentation depths of 750–800 nm.

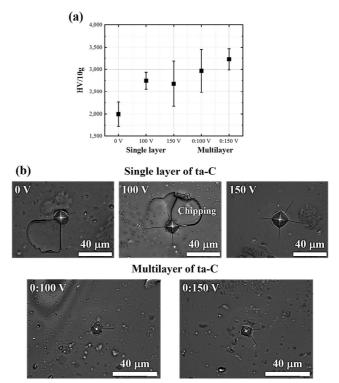
Meanwhile, for the multilayer coatings, cracks started to form at an indentation depth of 700–800 nm, as

shown in Fig. 5(b). However, crack propagation was hindered by the presence of multiple layers.

Figure 6 shows the condition of the surface after indentation with a higher normal load of 10 g using the micro Vicker indentation method. The nano-indentation results indicate that the hardness of the single layer coatings was consistently lower than that of the multilayer coatings, and a tendency to increase and then decrease with increasing  $V_b$  was exhibited. Although the hardness values could not be obtained based on the micro Vicker indentation, the crack propagation on the surface can be obtained. As shown in the variation in  $H^3/E^2$  values in Fig. 5, the crack length was proportional to the resistance to plastic deformation. This means that the multilayer coatings exhibited better resistance to crack propagation than the single layer coatings. This result is consistent with that obtained for the nanohardness.

## 3.3 Adhesion strength of ta-C coatings

To determine the adhesion strength of the multilayer coatings, scratch tests were performed. Figure 7(a) shows the scratch test results for all ta-C coatings. For the multilayer coatings, adhesion strengths of 29 and



**Fig. 6** Microhardness values for single layer and multilayer of ta-C coatings and optical micrographs of each sample.

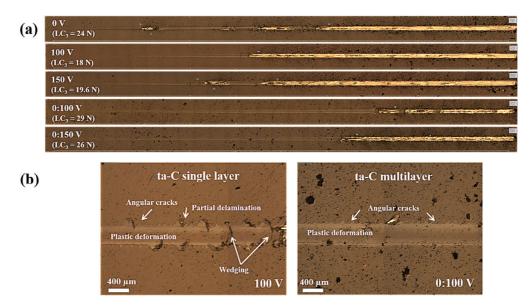


Fig. 7 (a) Scratch tracks and optical observation of (b) single layer (100 V) and multilayer (0:100 V) ta-C coatings.

26 N were obtained when the coatings were deposited at  $V_b$  of 0:100 and 0:150 V, respectively. In the case of single layer coatings, adhesion strengths of 24, 18, and 19.6 N were obtained for  $V_b$  values of 0, 100, and 150 V, respectively. Based on these results, it can be concluded that the structure of the coatings significantly affected the adhesion strength. Single layer coatings typically have lower  $H^3/E^2$  values than multilayer coatings. Therefore, delamination, crack propagation, angular cracking, partial delamination, and wedging occurred at the beginning of the scratch test (Fig. 7(b)), all of which were attributable to the brittleness of the single layer ta-C coating.

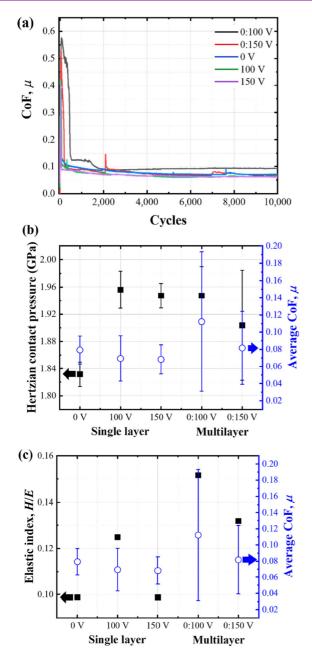
Although one expects the multilayer coatings to be more brittle than the single layer coatings because of their high hardness values, only angular cracks were detected in the scratch patterns, as shown in Fig. 7(b). These results show that the multilayer coating structure can prevent crack propagation through internal stress relaxation at the soft ta-C layer. Because the multilayer coating was formed using relatively soft ta-C layers with a substrate bias of 0 V, we inferred that the soft layers might contribute positively to the tribological behavior because of the absence of partial delamination and wedging.

These results show that the use of a multilayer structure for the deposition of ta-C coatings through bias control results in a significant inhibition in crack propagation by internal stress release.

## 3.4 Tribological properties of ta-C coatings

Figures 8 and 9 show the variation in the friction coefficient (CoF), average CoF, and wear rate after 10,000 rotation cycles of the ta-C coatings. For single layer coatings, the CoF decreased from 0.079 to 0.068, whereas the wear rate increased from  $3.052 \times 10^{-7}$  to  $11.024 \times 10^{-7} \text{ mm}^3/(\text{m}\cdot\text{N})$  with increasing  $V_b$ . Even though the single layer coatings deposited with 0 and 150 V indicated similar hardness and  $H^3/E^2$  values, their wear resistances differed significantly. This is attributable to the differences in the internal stresses, which may be affected by the applied  $V_b$ . However, in the case of multilayer coatings, the CoFs were in the range between 0.082 and 0.112, which were slightly higher than that of the single layer coatings. This is due to the presence of a higher ratio of the hard layers, such as the coatings deposited with  $V_b$  of 100 and 150 V. Although the CoF was slightly higher, the multilayer coating deposited with a  $V_b$  of 0:100 V had a wear rate of  $2.019 \times 10^{-7}$  mm<sup>3</sup>/(m·N). A decrease by 34% in comparison with the single layer coatings prepared under similar conditions was indicated. These results were consistent with the increase in the resistance to plastic deformation and the increase in the  $H^3/E^2$ values.

Erdemir et al. [25] reported that the reciprocal ratio of the Hertz contact pressure to the shear strength affected the CoF. However, in this study, because the



**Fig. 8** (a) CoF behaviors are against  $Al_2O_3$  ball; (b) average CoF is shown in blue, and Hertzian contact pressure is shown in black; (c) average CoF is shown in blue, and elastic index is shown in black.

plasticity index was 6–10, the contact state belonged to the elastic–plastic contact, and the Hertzian contact pressure and CoF were not correlated. However, Waghmare et al. [26, 27] reported that the elasticity index of *H/E* should be considered along with the Hertz contact pressure. Therefore, as shown in Figs. 8(b) and 8(c), the elasticity index and average CoF indicated reliable results.

In elastic contact, wear occurs at ta-C as adhesive and abrasive wear, which is proportional to the contact area [27]. However, because the tribology test involved a plastic/elastic contact area, the wear rate should exhibit a tendency to be inversely proportional to the hardness. However, in this study, it was difficult to describe the wear using a general wear definition because cracks occurred owing to the high contact load. Therefore, we explained and verified the crack based on the  $H^3/E^2$  ratio and the indentation test. The wear resistance of the single layer and multilayer ta-C coatings under a high normal load of 30 N was analyzed by analyzing the mechanical properties such as hardness, elastic modulus, adhesion strength, friction, and wear properties. Duminica et al. [7] reported that coating classification performance is based on an index (I) coupling, the mechanical and tribological properties of diamond-like carbon (DLC) coatings. Furthermore, they reported that the ratios  $H^3/E^2$  and  $F_{1c3}/\mu$  represent resistances to plastic deformation and wear in that index, respectively. Fig is the load at which the ta-C coating was completely removed from the scratch track. Figure 9 shows that the  $H^3/E^2$ values obtained from our experiments correlated well with the wear rate of the ta-C coatings. The wear rate decreased when the value of  $H^3/E^2$  was higher for the coating, except for the coating deposited with a substrate bias of 0 V. Coatings deposited at a  $V_b$  of 0 V exhibited poor mechanical properties compared with the other coatings. The tribology test was conducted at a load of 30 N, and a high contact load was applied to the coating surface. As shown in Fig. 10, all of the ta-C coatings deposited in a single

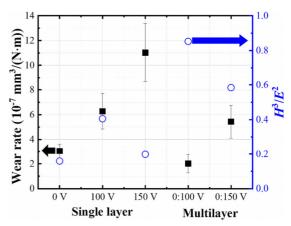


Fig. 9 Wear rate and  $H^3/E^2$  value vs. applied substrate bias voltages.

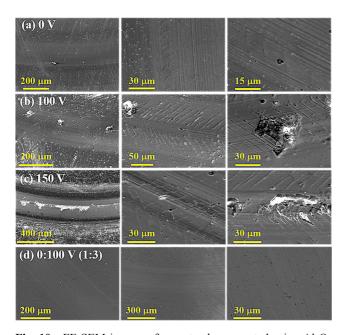


Fig. 10 FE-SEM images of wear tracks generated using Al<sub>2</sub>O<sub>3</sub> ball under applied normal load of 30 N on single layer ta-C coatings deposited with substrate bias voltages of (a) 0, (b) 100, and (c) 150 V, as well as on multilayer ta-C coatings deposited with substrate bias voltage of (d) 0:100 V.

layer structure possessed cracks; however, in the case of 0 V, cracks occurred, although delamination did not occur. Nonetheless, owing to the three-body abrasion caused by the delamination in the ta-C coating arising from the high residual stress, the single layers of 100 and 150 V exhibited high wear. Therefore, in the case of 0 V, even though  $H^3/E^2$  was low, the residual stress was low as well; therefore, the wear rate was considered to be lower than that of the other single layer coatings. Considering our wear rate results, a high  $H^3/E^2$  value indicates good wear resistance of the multilayer coating and provides information for evaluating the tribological performances of coatings.

Figure 10 shows the FE-SEM images of the wear track on the ta-C coating against the Al<sub>2</sub>O<sub>3</sub> ball. We observed that for the single layer coatings, with increasing  $V_b$ , both the shape and size of the crack on the worn surface increased with fatigue failure. When  $V_{\rm b}$  increased, the size of the wear track increased, showing fatigue failure. In addition, the angular cracks observed in the scratch tests of the single layer ta-C coating performed at  $V_b$  values of 100 and 150 V were observed in the wear track. Moreover, the wear track of the single layer coating deposited at 100 V showed cracks and intermittent holes, which exposed the

underlying Ti interlayer and HSS substrate. These properties were verified via EDS analysis, as shown in Fig. 11.

The multilayer coatings exhibited smooth wear tracks, and no cracks were observed on the surface. We discovered that the multilayer structure halted crack propagation at the contact surface under high normal loads. The  $H^3/E^2$  ratio related to the plastic deformation resistance is in good agreement with the wear resistance. As the fracture toughness increased, the elasticity of the coatings increased as well. These findings were consistent with the results obtained from the nanoindentation experiments (Fig. 5).

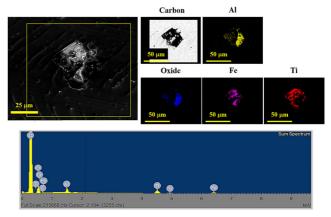


Fig. 11 EDS analysis of single layer ta-C coating deposited using substrate.

## **Conclusions**

We proposed a multilayered deposition method of ta-C coating to enhance mechanical properties such as hardness and wear resistance. Characterization of the multilayered structure of ta-C obtained via a cyclic deposition of soft and hard layers, indicating that its mechanical properties such as hardness, elastic modulus, H³/E² value, adhesion strength, friction, and wear resistance can be modified significantly. The conclusions of the study are as follows:

1) A multilayered ta-C coating with a thickness of 2.0 µm composed of alternating layers of soft and hard layers was successfully fabricated using a FCVA with an alternating substrate bias voltage. FE-SEM analysis revealed that the soft-to-hard layer thickness ratio was 1:3 with the actual thicknesses being 100 and 300-340 nm for the soft and hard layers, respectively.

- 2) Characterization of the mechanical properties of the coatings, such as their hardness, elastic modulus, and  $H^3/E^2$  ratio indicated that the multilayer ta-C coatings performed better in comparison with the single layer ta-C coatings. In particular, the  $H^3/E^2$  ratio increased significantly from 0.159 to 0.852 as the alternating substrate bias increased from 0 to 100 V.
- 3) Multilayer coatings with higher  $H^3/E^2$  ratios showed significantly improved adhesion strength and wear performance under a normal load of 30 N owing to the impeded crack propagation through internal stress relaxation.

# Acknowledgements

This work was supported by the Fundamental Research Program of the Korea Institute of Materials Science (KIMS/PNK7000) and the Fundamental R&D Program of the Ministry of Science, Information & Communication Technology (ICT), and Future Planning in Republic of Korea.

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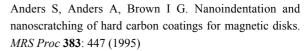
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