Effect of Plasma Nitriding on Multilayer Diamond-Like Carbon Films

Yusei Ogawa[1,*1] and Akio Nishimoto[2]

[1]*Department of Chemistry and Materials Engineering, Graduate School of Science and Engineering, Kansai University, Osaka 564-8680, Japan*
[2]*Department of Chemistry and Materials Engineering, Faculty of Chemistry, Materials and Bioengineering, Kansai University, Osaka 564-8680, Japan*

In this study, after strengthening a stainless steel surface by forming a nitrided layer (S phase) on the surface through S-DCPN treatment, multilayer DLC films with different numbers of layers are deposited to investigate the effects of the number of layers on the mechanical properties of stainless steel. An austenitic stainless steel SUS304 is used as the sample, and S-DCPN treatment is performed at a treatment temperature of 673 K, treatment time of 4 h and 16 h, gas pressure of 200 Pa, and a treatment gas composition of N_2:H_2 = 3:1. After S-DCPN treatment, DLC (a-C:H) films with different multilayer structures are deposited using a high-frequency plasma CVD system, to achieve a total film thickness of 1.2 μm. The results of this study show that the S-DCPN treatment strengthens the surface of the sample and improves the adhesion and durability of the DLC film. Also, with 673 K – 4 h nitriding + DLC, the sliding distance increased significantly compared to the untreated substrate, and 4-layers coating had the best durability.

Keywords: *diamond-like carbon films, s phase, S-DCPN, adhesion and durability, duplex process*

1. Introduction

Physical vapor deposition (PVD) and chemical vapor deposition (CVD) form hard coatings on surfaces, which are endowed with various functions that depend on the sample to be formed. Among the different surface modifications, diamond-like carbon (DLC) films have attracted much attention because they reduce the wear on sliding parts and improve the wear resistance, thereby extending the service life of parts. However, when forming a hard film, the adhesion between the film and sample is affected by the physical properties such as the hardness and coefficient of thermal expansion. Therefore, efforts are being made to improve this consistency by introducing multiple intermediate layers and varying the number of layers to reduce the residual stress in the DLC film, and through treatments such as nitriding and carburizing to harden the substrate by forming a hard film. Active screen plasma nitriding (ASPN) is a nitriding method that eliminates the edge effect by insulating the sample during the process. However, during nitriding, deposits cover the sample and slow down the nitriding rate. Therefore, an improved nitriding method, S-DCPN: direct current plasma nitriding with screen, in which a voltage is applied to both the sample and screen during ASPN to remove the deposits through sputtering and increase the nitriding speed has been reported[1]. Although a combination of ASPN and DLC coating is performed often, there have been very few reports on a duplex treatment with S-DCPN[2]. In this study, after strengthening a stainless steel surface by forming a nitrided layer (S phase) on the surface through S-DCPN treatment, multilayer DLC films with different numbers of layers are deposited to investigate the effects of the number of layers on the mechanical properties of stainless steel.

2. Experimental procedure

An austenitic stainless steel SUS304 is used as the sample, and S-DCPN treatment is performed at a treatment temperature of 673 K, treatment time of 4 h and 16 h, gas pressure of 200 Pa, and a treatment gas composition of N_2:H_2 = 3:1. After S-DCPN treatment, DLC (a-C:H) films with different multilayer structures are deposited using a high-frequency plasma CVD system, to achieve a total film thickness of 1.2 μm. After Ar bombardment to clean the substrate surface, the Si-DLC intermediate layer was deposited with a mixture of $Si(CH_3)_4$ and CH_4 gas, followed by the DLC film with CH_4. The total processing time was set to 30 min for the Si-DLC intermediate layer and 60 min for the DLC film using CH_4 gas. The deposition conditions were as follows: 2 layers of Si-DLC : DLC = 600 nm : 600 nm, 4 layers of 300 nm : 300 nm repeated twice, and 6 layers of 200 nm : 200 nm repeated 3 times. A schematic diagram of the film structures is shown in Figure 1. The deposition temperature was 443 K in both cases. Various properties of the treated samples were evaluated by X-ray diffraction, surface roughness, hardness, cross-sectional microstructure observation, glow discharge emission spectroscopy (GD-OES), Rockwell indentation, friction and wear, delamination test, and scratching test.

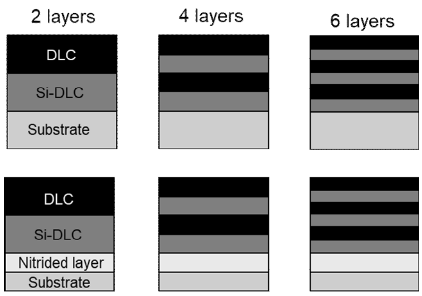

Fig. 1 Schematic diagram of multilayer DLC.

3. Results and discussion

3.1 Vickers hardness test

Figure 2 shows the results of hardness measurement of the cross sections after S-DCPN treatment. The hardness of the sample surface increased significantly after S-DCPN treatment. The thickness of the nitrided layer observed in the cross-sectional microstructures and the nitrogen-enriched area by GD-OES were consistent.

*1 Graduate Student, Kansai University

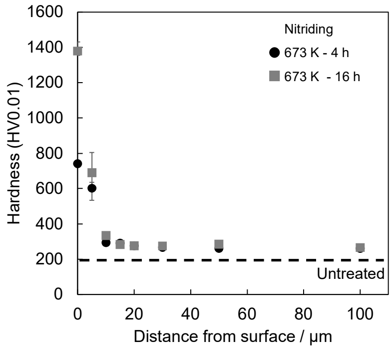

Fig. 2 Surface and cross-sectional hardness of SUS304 treated by S-DCPN.

3.2 Surface roughness

The results of surface roughness measurements of the S-DCPN-treated and DLC-deposited samples are shown in Fig. 3. As a result, the surface roughness after nitriding was larger than that of the untreated substrate. This is thought to be due to accelerated sputtering on the sample surface during the nitriding process. The surface roughness after DLC deposition was almost the same as that before nitriding for all samples. This is thought to reflect the surface properties of the substrate directly on the DLC film surface.

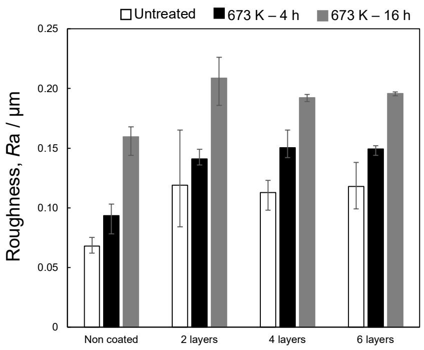

Fig. 3 Surface roughness of SUS304 treated by S-DCPN.

3.3 Rockwell indentation test

Figure 4 shows the results of the Rockwell indentation test for the sample with DLC film deposited after S-DCPN treatment. Figure 4 shows that the sample with the DLC film deposited on the untreated substrate showed partial peeling of the film around the indentation, which was judged to be HF 2. On the other hand, the S-DCPN-treated samples all showed good adhesion with HF 1. This is considered to be due to the strengthening of the substrate surface by S-DCPN treatment as a pretreatment.

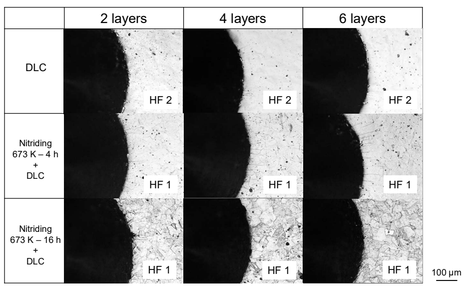

Fig. 4 Appearance of the region around indentation of SUS304 after Rockwell indentation test.

3.4 Friction and wear test

Figure 5 shows the results of the delamination test of a sample with a DLC film deposited on it. The durability of the film was evaluated based on the sliding distance of the ball from the top surface of the film to the substrate when the coefficient of friction curve reached μ = 0.3. Figure 5 shows that the sliding distance increased with increasing the number of layers in the untreated material coated with DLC. The sliding distance of 673 K – 4 h nitriding + DLC increased significantly compared to the untreated material, and the four layers were the most durable. 673 K – 16 h nitriding + DLC showed a decrease in sliding distance compared to the 4-hour treated material. This may be due to the increase in surface roughness caused by the longer nitriding treatment time, which adversely affected the durability of the film.

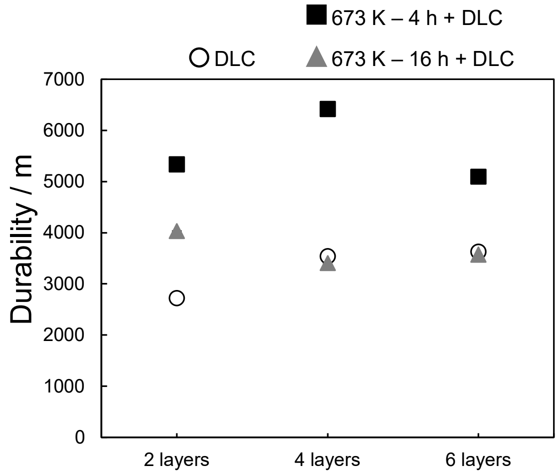

Fig. 5 Sliding distance after friction and wear test.

4. Conclusions

The following results were obtained from an investigation of the effects of the number of layers of multi-layer DLC films on the mechanical properties of austenitic stainless steel SUS304 after S-DCPN treatment and DLC film deposition by plasma CVD.

(1) S-DCPN treatment strengthens the substrate surface and improves the adhesion and durability of the DLC film.

(2) 673 K – 4 h nitriding + DLC coating significantly increased the sliding distance compared to the untreated material, and 4-layers coating had the best durability.

References

1) A. Nishimoto, T. Fukube and T. Tanaka : Mater. Trans. **57** (2016) 1811-1815.
2) A. Nishimoto, R. Amano and T. Tamiya : Appl. Surf. Sci. Adv. **6** (2021) 100129.