Effect of DLC and Si-DLC Films Deposited on Engineering Plastics on Tribological Properties

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In this study, diamond-like carbon (DLC) with different source material gases was deposited on engineering plastics by plasma enhanced chemical vapor deposition (CVD), and effect of DLC Films on tribological properties were investigated. Results have revealed that the lower the ratio of elements that inhibit carbon bonding in the source gas used for film deposition, the harder the DLC film becomes and the better its properties are under friction and wear conditions.

Keywords: engineering plastics, plasma-enhanced CVD, DLC, tribological properties

1. Introduction

As global environmental awareness grows, it is expected that parts will increasingly be replaced with engineering plastic materials from metallic materials to reduce fuel consumption in automobiles and other transportation equipment and improve portability of home appliances and various devices by reducing their weight. Hence, there is a strong social need to improve wear and friction resistance in engineering plastic materials through surface modification treatments¹⁾. DLC is particularly useful for improving the friction and wear properties of sliding parts because of its high hardness, low friction, chemical stability, and excellent seizure resistance²⁾. However, one drawback of DLC is the thermal stress caused by the difference in the thermal expansion coefficient between the film and the substrate material and the internal stress incorporated into the film during its formation, which could reach as high as several GPa. This results in a high elastic strain energy, which reduces adhesion³⁾. Therefore, research is being conducted to improve adhesion by reducing thermal stress through the introduction of an intermediate layer with a thermal expansion coefficient between that of the DLC and the substrate material or by reducing internal stress through the introduction of a DLC intermediate layer containing metallic elements. In this laboratory, research has been underway to reduce internal stress and improve adhesion by adding Si into DLC. In this study, DLC and Si-DLC films were deposited on engineering plastics using the plasma CVD method, and the effects on adhesion, mechanical properties, and friction and wear properties of the plastic substrates and DLC films were investigated.

2. Experimental procedure

Using polyetheretherketone (PEEK) as the substrate material, a DLC film was deposited on each sample using a high-frequency plasma CVD system to achieve a film thickness of 1 μ m. After Ar bombardment to clean the substrate surface, DLC films were deposited using CH₄ or C₂H₂ as the source gas, and Si-DLC films were deposited using (CH₃)₄Si and CH₄ or (CH₃)₄Si and CH₄ or (CH₃)₄Si and C₂H₂ gases.

The processing temperature was 298 K for all films. Visual observation, surface roughness test, film thickness measurement, Rockwell indentation test, friction and wear test, Raman spectroscopic analysis, GD-OES, and nano indentation test were performed on the treated samples.

3. Result and discussion

3.1 Rockwell indentation test

The Rockwell indentation test was performed to evaluate the adhesion of the entire films. In the Rockwell indentation test, the delamination around the indentation is observed and judged by HF 1 to HF 6 based on the German standard VDI3198. If there is little delaminating and cracking around the indentation, the adhesion is considered good, and if there are large delaminating and cracks crossing, the adhesion is considered poor. The results are shown in Figure 1. Under all conditions, HF 1 and HF 2 showed excellent adhesion; if there is a difference in hardness between DLC and the substrate as a property of DLC, the adhesion will be poor because of the difference in the strain process during deformation. In this study, the adhesion was excellent even though the film was deposited on a resin material with much lower hardness than DLC. This is thought to be due to the reaction at the adhesive interface between the DLC and the substrate.



Fig. 1 Rockwell indentation of various Si-DLC or DLC.

3.2 Friction and wear test

Figure 2 shows the friction coefficient during sliding and the distance required for delaminating for each sample. The test was continued until the friction coefficient reached that of PEEK (approximately 0.4), at which point the film was considered to have peeled off. Compared to the Si-DLC film, the DLC film has a lower coefficient of friction than the Si-DLC film, and delaminating was observed at a much longer

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sliding distance. Compared by gas type, the sample deposited with C_2H_2 gas had a lower coefficient of friction and a longer sliding distance than the sample deposited with CH₄ gas. It was also found that the lower the coefficient of friction, the longer the sliding distance to delamination.



Fig. 2 Delamination distance and friction coefficient of Si-DLC or DLC.

3.3 Nano indentation test

The results of the nanoindentation test are shown in Figure 3. Since the film thickness of both specimens was the same, the test was conducted with the indentation load set to 0.8 mN so that the indentation depth was less than 1/10 of the film thickness. When compared by gas type, the DLC film with C₂H₂ gas showed greater nano hardness than the DLC film with CH₄ gas. This is thought to be because CH₄ has a higher H content than C₂H₂, and H terminates a higher percentage of carbon-to-carbon bonds (C-C, C=C) in the DLC. The nano hardness and Young's modulus of the Si-DLC film were smaller than those of the DLC film. This is thought to be due to the presence of Si in addition to H as an element that terminates carbon bonds in Si-DLC, which increases the number of Si-C bonds. The nano-hardness of each film was found to be positively correlated with the delamination distance obtained in the friction and wear tests and negatively correlated with the coefficient of friction. These results suggest that the hardness of the films has a significant effect on the tribological properties of DLC deposited on plastics.



Fig. 3 Film hardness and Young's modulus of Si-DLC or DLC.

3.4 Raman spectrometry

Figure 4 shows the positions of the G peaks obtained by peak separation of the Raman spectra, where the G peaks originate from vibrations of all sp² bonds, both on the chain and in the ring, and the larger the value, the larger the sp^2 bonding ratio. Figure 4 shows that the DLC sample has a larger ratio of sp² bonds than the Si-DLC sample. This is thought to be because the carbon bonds are terminated by the H and Si contained in the TMS gas used for Si-DLC deposition, making it difficult to form a graphite structure. In addition, a comparison by gas showed that the sample deposited using C₂H₂ gas had a larger ratio of sp² bonds than that deposited using CH₄ gas. This is thought to be because the H content in CH₄ gas is larger than that in C₂H₂ gas and H terminates the bonds between carbons more frequently. The larger the sp² bonding ratio, the larger the delamination distance in the friction and wear tests. These results suggest that the degree of graphitization of the films affects the tribological properties.



Fig. 4 G peak position of PEEK coated by DLC or Si-DLC.

4. Conclusions

To investigate the effect of DLC and Si-DLC films deposited on engineering plastics on tribological properties, Si-DLC and DLC films were deposited using a highfrequency plasma system. The results revealed as follows: (1) In both conditions, adhesion was good despite the large difference in hardness between the films and substrates. (2) The larger the ratio of elements that inhibit carbon bonding in the source gas, the more graphitized and harder the film becomes. (3) The hardness of the film and the degree of graphitization play a significant role in the tribological properties of DLC deposited on plastics.

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