

Adhesion Strength and Optical Transparency of DLC Coatings on Polycarbonate

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In this study, we investigated the adhesion strength and optical transparency of a-C, a-C:H, a-C:H:Si and SiO₂ films deposited on polycarbonate. The a-C and SiO₂ films were deposited by ECR-type ion beam sputtering system (ECR-type IBS), and the a-C:H and a-C:H:Si films were prepared with bipolar-type plasma based ion implantation (bipolar-type PBII). It is observed that the friction signal of the polycarbonate decreases due to the diamond-like carbon (DLC) coating, and the a-C:H:Si coating shows the lowest friction force. The a-C:H:Si film on the polycarbonate shows higher critical load, i.e., higher adhesion strength, compared to those of the a-C, a-C:H and SiO₂ coatings. The transmittance increases due to the Si-addition to the a-C:H film in a wavelength range of 400 – 800 nm (visible range), whereas decreases in a wavelength range of 200 – 400 nm (UV range). The a-C:H:Si coating exhibits a larger optical band gap compared to those of other coatings due to highly sp³ dominant bonding structure. The a-C:H:Si is effective protective-coating for the polycarbonate due to its lower friction higher adhesion strength, higher transmittance in the VIS range and better UV protection ability compared to those of other coatings.

Key words: diamond-like carbon, polycarbonate, adhesion strength, optical transparency, bipolar-type PBII&D

1. INTRODUCTION

Polycarbonate is used in various applications such as automobile headlamps, compact discs, safety shield of window, ophthalmic lenses due to its excellent impact strength, stiffness and optical transparency. However, the limitations according to low scratch resistance and weakness for ultraviolet (UV) radiation restrict their practical application. Ceramic coatings can be applied on polycarbonate to protect them against scratch and UV radiation. Though several coatings such as zinc oxide, silicon oxide, silicon nitride and titanium oxide on polycarbonate have been reported [1-4], we consider diamond-like carbon (DLC) coatings offer the best protection performance for the polycarbonate due to its high hardness and low friction, and chemical inertness.

In this study, the DLC coatings were deposited on the polycarbonate using ion beam sputtering and plasma based ion implantation, and their scratch resistance and optical properties were investigated.

2. EXPERIMENTS

2.1 Deposition of the a-C, a-C:H and a-C:H:Si films

The a-C films were deposited on the polycarbonate and quartz glass substrates using an ECR-type ion beam sputter (Ellionix, model EIS-200ER) with a graphite target, which is shown in Fig.1(a). For the comparison purpose, the SiO₂ films were also deposited using the ion beam sputter with a SiO₂ target. The deposition conditions are as follows; acceleration voltage of Ar⁺ ion beam = 2000 V, microwave

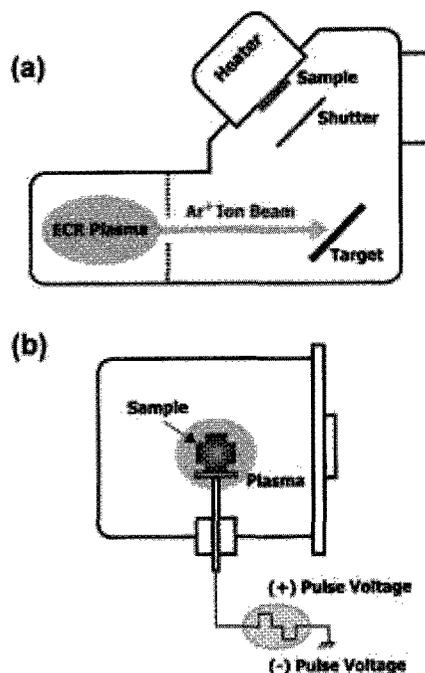


Fig. 1. (a) ECR-type ion beam sputtering system (ECR-type IBS) and (b) bi-polar type plasma based ion implantation and deposition system (bipolar-type PBII&D) used in this study.

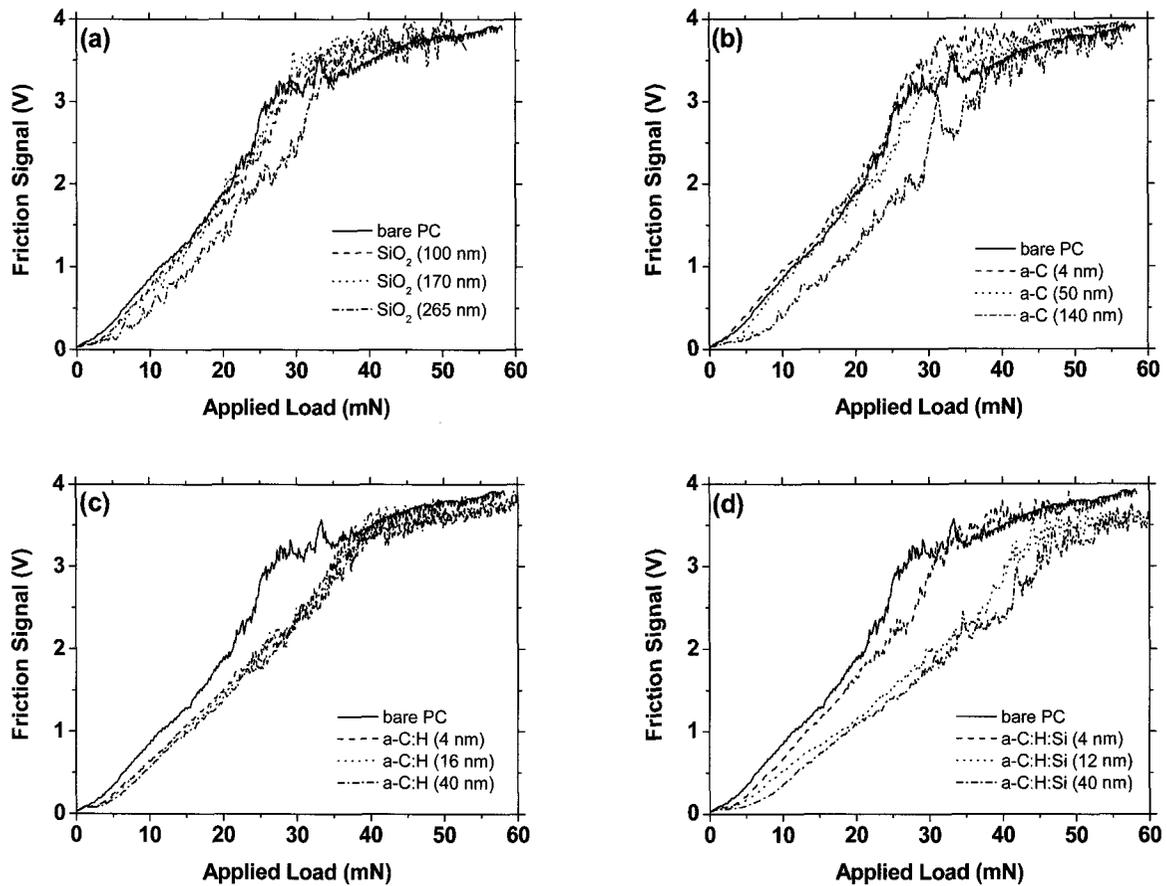


Fig.2 Scratch profiles of (a) SiO₂, (b) a-C, (c) a-C:H and (d) a-C:H:Si coatings deposited on the polycarbonate.

power = 100W, deposition pressure = 8×10^{-3} Pa.

A bipolar-type PBII&D system (Fig.1(b)) [5] was used for the deposition of a-C:H and a-C:H:Si films on the polycarbonate and quartz glass substrates. The deposition conditions of the films are as follows; precursor gases: toluene for a-C:H film, a mixture of tetramethylsilane and toluene for a-C:H:Si film, positive and negative pulse voltages: +2.0 and -5.0 kV, pulse frequency: 4 kHz, pulse duration: 5 μ s. The deposition temperature was low enough not to deform the polycarbonate.

The composition of the deposited films were measured by x-ray photoelectron spectroscopy (XPS) (Sigma Probe, model Thermo VG) and elastic recoil detection analysis (ERDA). The thickness of the deposited films was determined using deposition rates.

2.2 Evaluation of the adhesion strength

The adhesion strength of the coatings deposited on the

Table I. The compositions of the deposited DLC coatings measured with XPS and ERDA.

Deposition Method	Sample Name	C (at.%)	H (at.%)	Si (at.%)
IBS	a-C	100	-	-
PBII	a-C:H	81	19	-
PBII	a-C:H:Si	63	20	17

polycarbonate was evaluated with a micro-scratch tester (Rhesca, model CSR-02) by measuring the critical load. Scratch tester is used for evaluating the bonding strength between the thin coating and substrate as well as mechanical durability of the film surface, by pressing and scratching the spherical diamond indenter against the film surface with gradually increasing the normal load. When a thin coating is destroyed at a normal load (i.e., critical load), the friction signal is abruptly changed [6]. The radius curvature of the diamond tip, the load rate and the scratching angle were 5 μ m, 34.8 mN/mm and 2 degrees, respectively.

2.3 Measurements of the optical properties

The transmittance of the deposited coatings on the polycarbonate and quartz glass was measured with UV/VIS spectrophotometer (Jasco, model V-530) in the wavelength range of 200 – 800 nm. The scanning rate was 1.67 nm/s.

3. RESULTS AND DISCUSSION

3.1 Composition of the deposited DLC coatings

Table I shows the composition of the DLC coatings. It is observed that the hydrogen contents of the a-C:H and a-C:H:Si are almost same. It means that the incorporated Si atoms in the DLC film are substituted for the C atoms. Substitution of Si atoms for sp^2 or sp^3 hybridized C atoms makes increase the sp^3 hybridized bonds in the DLC film because Si atoms usually make sp^3 hybridized bonding. It

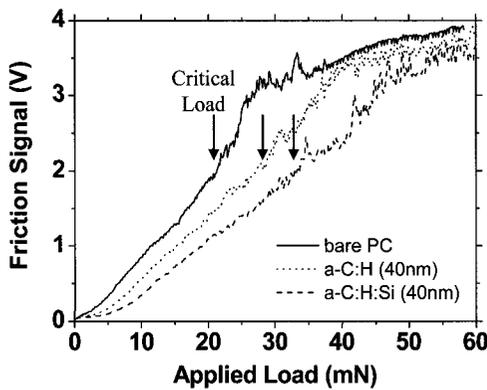


Fig.3 Micro-scratch profiles of 40-nm-thick a-C:H and a-C:H:Si coatings deposited on the polycarbonate.

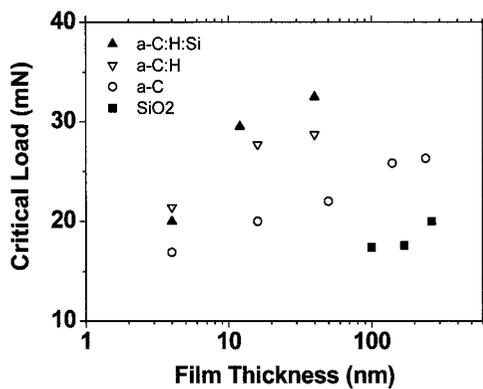


Fig.4 Critical loads of the coatings deposited on the polycarbonate, which were obtained from the scratch profiles.

has been reported that the Si incorporation in the DLC films increases the sp^3 bonding in the film [7,8].

3.2 Adhesion strength of the deposited films on the polycarbonate

Figure 2 shows the scratch profiles of the a-C, a-C:H, a-C:H:Si and SiO₂ coatings on the polycarbonate. For the comparison, the scratch profile of the bare polycarbonate is also shown. In Fig.2, slope of the scratch profile means friction coefficient. It is observed that SiO₂ coatings (or a-C coating) thinner than 265 nm (or 140 nm) do not contribute to reduce the friction of bare polycarbonate. On the other hand, 4-nm-thick a-C:H and a-C:H:Si coatings effectively reduce the friction of the polycarbonate, and the thicker film thickness is, the lower friction coefficient. Figure 3 shows the scratch profiles of 40-nm-thick a-C:H and a-C:H:Si coatings on the polycarbonate. It is observed that the a-C:H:Si coating shows a lower friction coefficient compared to that of the a-C:H coating.

The critical loads of the coatings deposited on the polycarbonate can be obtained from the scratch profiles shown in Fig.2. As shown in Fig.4, the critical load increases with increasing film thickness. The a-C and SiO₂ coatings deposited using IBS exhibit lower critical loads than those of the a-C:H and a-C:H:Si coatings deposited using PBII because in the PBII the carbon ions with higher kinetic energy reach on the polycarbonate surface. The a-C:H:Si

coating show a higher critical load compared to that of the a-C:H coatings due to lower internal stress of the a-C:H:Si coating [9]. On the other hand, the critical loads of 4-nm-thick a-C:H:Si and a-C:H coatings are comparable. It means that the effect of the internal stress on the critical load decreases with the decreasing film thickness.

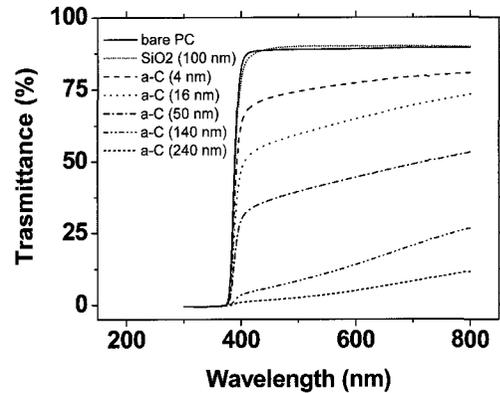


Fig.5 UV/VIS transmittance spectra of a-C films deposited on the polycarbonate. The thickness of a-C films was changed up to 240 nm.

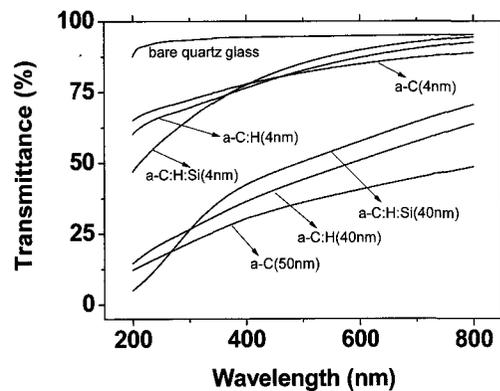


Fig.6 UV/VIS transmittance spectra of the a-C, a-C:H and a-C:H:Si coatings deposited on the quartz glass.

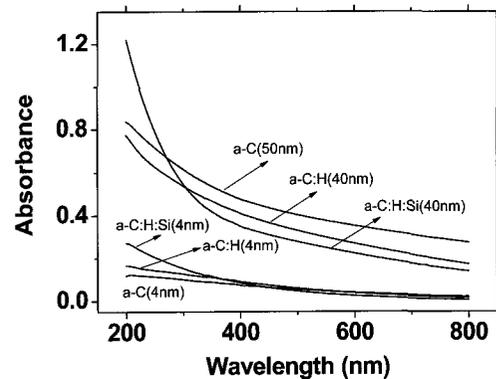


Fig.7 UV/VIS absorbance spectra of the a-C, a-C:H and a-C:H:Si coatings deposited on the quartz glass.

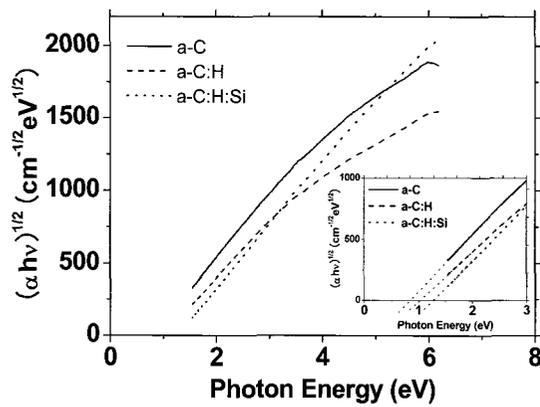


Fig.8 Absorption coefficient of a-C, a-C:H and a-C:H:Si films.

Table II. Optical band gap of the DLC films.

Optical band gap (eV)	
a-C	0.84
a-C:H	1.03
a-C:H:Si	1.27

3.3 Optical properties of the deposited films

Figure 5 shows UV/VIS transmittance spectra of the a-C films deposited on the polycarbonate in the wavelength range of 200-800 nm. For the comparison, the spectrum of SiO₂ film with a thickness of 100 nm is also shown in the figure. It is apparent that the transmittance of the a-C coatings decreases with the increasing film thickness. The transmittance of the polycarbonate with 50-nm-thick a-C coating is reduced to below 50 %. The transmittance of the SiO₂ coating is comparable to that of the bare polycarbonate.

The a-C, a-C:H and a-C:H:Si coatings were deposited on quartz glass to investigate the optical properties of the coatings in the UV range. The UV/VIS transmittance and absorbance spectra of the coatings were shown in Fig. 6 and Fig.7, respectively. The absorbance spectra were obtained from the transmittance data using the following relationship; $I = I_0 \exp(-A)$, where I_0 is the intensity of the incident light and I is the intensity of the light coming out of the sample, and A is the absorbance. It is observed that the a-C:H:Si coating shows better optical transparency in the VIS range and higher UV protection ability.

The optical band gap (E_{opt}) of the DLC coating can be obtained through the Tauc plot. The intercept of the linear

portion of the $(\alpha E)^{0.5}$ plot on the photon energy (E) axis gives the Tauc band gap (right inset of Fig.8), where $\alpha (=A/d)$ is the absorption coefficient of the coating and d is the thickness of coating [10]. The obtained band gap for 4-nm-thick DLC coatings are shown in Table II. The band gap of the a-C coating increases due to hydrogen or hydrogen and silicon incorporation in the carbon network, indicating a compositional change from less sp^3 dominant bonding structure toward highly sp^3 dominant bonding structure[11].

4. SUMMARY

The adhesion strength and optical transparency of a-C, a-C:H, a-C:H:Si and SiO₂ films deposited on the polycarbonate were investigated. The a-C:H:Si is effective protective-coating for the polycarbonate due to its lower friction, higher adhesion strength, higher transmittance in the VIS range and better UV protection ability compared to those of other coatings.

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