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Effect of sputtering pressure on the surface topography, structure, wettability and tribological performance of DLC films coated on rubber by magnetron sputtering

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ABSTRACT

Diamond-like carbon (DLC) films were deposited on the nitrile butadiene rubber substrates by magnetron sputtering. The effects of the Ar sputtering pressure (SP) on the surface topography, structure, wettability and tribological performance of the DLC films were investigated. The DLC films' morphology as a function of SP was studied by atomic force microscopy, and their structural properties were investigated using Raman spectroscopy and FTIR. The Raman results revealed that a high SP contribute to forming sp^3 bonds. The surface energy was calculated by the Owens-Wendt-Rabel-Kaelble method, and it peaked at a SP of 1.4 Pa. Tribological tests were performed using a ball-on-disc tribometry at a fixed load of 0.3 N under ambient conditions, which revealed that the tribological characteristics of DLC films can be tuned by varying the SP. The results showed that the DLC films prepared under 1.4 Pa SP exhibited enhanced triboperformance, based on a stable coefficient of friction lower than 0.25 during the whole test, which was very promising for practical tribological applications.

1. Introduction

In varied applications such as oil and gas production which include pipes, valves and seals, there are different degradation mechanisms including wear and wear-corrosion. For instance, rubber seals under dynamic working conditions suffer from severe wear because of high contact friction, leading to the failure service of bearings. Thus, robust protective coatings with low coefficient of friction and high wear resistance are highly required [1]. The development of diamond-like carbon (DLC) films has been the focus point in recent years due to its ultralow friction and wear performance under both lubricated and non-lubricated conditions [2–7].

DLC thin films have been prepared by various methods including pulsed laser deposition [8], ion beam sputtering [9], plasma enhanced chemical vapour deposition (PECVD) [10] and magnetron sputtering (MS) [11]. In this field, Lubwama et al. [12–15] used a combination of closed field unbalanced magnetron sputtering ion plating and PECVD with an Ar/C_4H_{10} plasma to deposit DLC and Si-DLC films with and without Si-C interlayer onto nitrile rubber at a low substrate bias of -30 V and investigated the triboperformance of DLC and Si-DLC films. Pei et al. [16] investigated the influence of the bias voltage of MS plasma cleaning and the pulse direct current of plasma chemical vapour

deposition on DLC films and provided generic design rules for the deposition of flexible and ultralow friction DLC films on rubber seals. MS, a kind of preparation method of DLC films, possesses many advantages such as conceptual simplicity, low cost, scalability, coating uniformity and surface smooth [17]. Considering the industrialization promotion of rubber seals, the direct current MS (DC-MS) is suitable for application due to its uncomplicated structure and efficient deposition.

Note that the process parameters of MS play a very important role in preparing films. Wang et al. [18] fabricated amorphous hydrogenated carbon films on silica substrates by MS and showed that the micro-structure, mechanical and tribological properties of the films strongly depend on the ion energy which can be controlled by the substrate bias voltage. N.W. Khun and E. Liu [19] investigated the influence of carbon sputtering power on the structures and properties of PtRuN-DLC films deposited on silica substrates using a DC-MS deposition system and found that the increasing carbon sputtering power significantly improved the wear performance of the films as a result of the enhanced adhesion strength of the films. Similarly, the pressure in sputtering process also affects the structure and performance of prepared films. For practical applications, the study of films on rubber is very crucial. Fortunately, the chemical composition of DLC mainly consists of C, suggesting a good compatibility with rubber materials. Besides, the

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rubber itself has a rough surface, which differentiates the properties of the films deposited on it from those on other substrates. Therefore, it is very worthy and challenging to study DLC films on the rubber.

In this study, DLC films were synthesized on nitrile butadiene rubber (NBR) using DC-MS under different sputtering pressure (SP) of Ar sputtering gas in vacuum chamber, which was tuned by regulating the opening size of the gate valve of molecular pump. Effects of pressure in vacuum chamber on the surface topography, structure, wettability and tribological properties of DLC films were investigated in details. The relationships among the surface topography, structure, and wettability of the films and their contributions to tribological behaviour with the variation of SP were also discussed.

2. Material and methods

DLC films were deposited on NBR by the DC-MS using Ar gas as the sputtering gas, and a high purity graphite (> 99.9%) target was fixed on the sputtering target as a carbon source. The rubber substrates were initially cleaned by two cleaning steps using a detergent solution and boiling water, respectively, in order to remove the dirt and the wax on the rubber and thus improve the film adhesion [20]. NBR is dried in vacuum oven after the cleanout. Prior to deposition, the substrates were sputter-cleaned for 10 min using Ar⁺ by an anode ion source with a bias voltage of −600 V, at a frequency of 40 kHz and duty ratio of 75%. And then several DLC films were produced under five different pressure in the vacuum chamber of 0.2 Pa, 0.6 Pa, 1.0 Pa, 1.4 Pa and 1.8 Pa, hereafter referred to as P2, P6, P10, P14 and P18, respectively. Five different values of pressure were obtained by controlling the opening size of gate valve. During the deposition, the rate of Ar gas flow and the target power were maintained at 30 sccm and 100 W respectively for whole experiments. Simultaneously, a pulse bias voltage of −150 V at a frequency of 40 kHz and duty ratio of 75% was applied on the substrate to further strengthen the adhesion. The deposition time was 25 min. No additional substrate heating was applied. Moreover, the cleaned uncoated-NBR substrates were also produced for comparisons in follow-up experiments.

The surface topography of the as-deposited films was revealed by adopting atomic force microscopy (AFM, Nanosurf AG) via a tapping mode with a silicon probe (Tap 190Al-G). Areas of 5 μm × 5 μm for each sample were employed for the general observation and detailed information. The structure and chemical bonding configurations of the DLC films were analysed using Raman spectroscopy with incident lights at wavelengths of 514.5 nm and FTIR (Spectrum One, Perkin-Elmer) recorded in the region of 2000–650 cm^{−1} respectively.

Static contact angle (CA) measurements were carried out at room temperature (RT) by the sessile drop method using droplet of 1 μL, which is sufficiently small to avoid the effect of the drop's impact due to its weight [21]. Finally, stationary CA was considered in the steady-state value at the end of the measurement after the 15th second. The same procedure was performed for all the materials and two liquids (diiodomethane and water), and the liquid parameters are shown in Table 1. For each combination of liquid and surface, the measurements were repeated five times in order to minimize the measurement uncertainty and the average value of the steady-state value and its standard deviation were calculated. The surface energy was calculated by the Owens-Wendt-Rabel-Kaelble (OWRK) method (Eq. (1)) [23]. In the method where θ is the average CA, surface energy is segmented into

dispersive component (γ_L^d) and polar component (γ_L^p), and subscript _s and _L represent solid and liquid, respectively.

$$\gamma_L(1 + \cos \theta) = 2(\gamma_s^d \gamma_L^d)^{1/2} + 2(\gamma_s^p \gamma_L^p)^{1/2} \quad (1)$$

The adhesion strength of the DLC films was evaluated using a micro-scratch tester (CETR-UMT, Bruker) with a diamond stylus of 12.5 μm in radius which was dragged down the films under progressive loading condition at RT. The scratch rate was 20 μm/s, and the load increased linearly from 10 to 200 mN in 40 s. The scratches tracks were observed using scanning electron microscopy (SEM) (Phenom ProX). The tribological properties of the samples were investigated using a ball-on-disk tribometer (CETR-UMT, Bruker) under dry conditions at RT. Each sample was tested against a tungsten carbide ball of Φ1.6 mm in a circular path of Φ9 mm at a speed of 100 rpm under a normal load of 0.3 N, and every trial lasted for 75 min. The surface profile of the samples was determined by a Dektak-XT surface profiler with a stylus force of 3 mg over a scanning range of 1 mm. A laser confocal microscope (VK-X200 series, KEYENCE) was used to observe wear tracks.

3. Results and discussion

Fig. 1 shows the AFM images of uncoated and DLC-coated NBR under various SP. An obvious difference is that carbon atom clusters only appeared on the coated NBR's surfaces rather than uncoated NBR' surface. Fig. 1 indicates that the surface roughness of NBR is 274.35 nm, while the lowest roughness can be as low as 41.03 nm provided NBR was coated by the DLC film. As for NBR, its surface is relatively rough and has many holes. Generally speaking, the surface roughness of NBR tends to reduce obviously after depositing DLC films on the NBR. It could be explained that sputtered carbon atoms are likely to accumulate in the holes embedded in the surface of NBR and fill the holes when DLC films are deposited on rough surfaces of NBR [24].

Closer observations indicated that the roughness tends to increase firstly and then decrease as SP increases. During the stage from P2 to P14, the main factor affecting surface roughness is the size of carbon atom clusters, which subjects to the energy of carbon atoms that will bombard on the surface of NBR substrate, and in fact, a high pressure will generate numerous Ar obstacle particles and reduce the energy of final carbon atoms in the scattering process. Nevertheless, once SP increased to 1.8 Pa, the roughness decreased. It is well known that the increase of SP correlates with the increase of the concentration of Ar ions in vacuum chamber, which renders the graphite target receive more sputtering than before and the sputtering yield is thus improved. Consequently, a mass of carbon atoms could be produced, and then the holes located on the surface of NBR could be filled considerably. In this case, the contribution of the cluster size to the surface roughness is less apparent than that of holes.

The chemical bonding of films has been studied by means of Raman spectroscopy. Fig. 2 shows the Raman spectra of DLC films synthesized under different SP. Generally, an asymmetric broad peak exists around 1500 cm^{−1} in the Raman spectroscopy of the DLC film. In order to obtain more information about carbon bond structures, the asymmetric Raman spectra were fitted with Gaussian line shapes, and the background was subtracted. The fitted two peaks, the G-peak around 1580 cm^{−1} and the D-peak around 1360 cm^{−1}, correspond to the stretching vibration of any pair of sp² bonds whether in chains or in aromatic rings and the breathing mode of those sp² bonds only in rings, not in chains, respectively [25]. Moreover, it is easy to find that the signal-noise ratios of DLC's Raman spectra become noticeably low when the SP is higher than 0.6 Pa as compared to 0.2 Pa. It is explained that a small amount of carbon film gives rise to a low intensity reflected in Raman spectra. Actually, the main reason for this phenomenon is the decrease of the average free path of atoms in the deposition process caused by the variations in pressure.

Much indirectly information with respect to carbon bonds, such as

Table 1
Surface tension components for liquids (293.15 K) [22].

Liquids	Total γ_L (mN/m)	Dispersion γ_L^d (mN/m)	Polar γ_L^p (mN/m)
Water	72.8	21.8	51.0
Diiodomethane	50.8	50.8	0.0

Fig. 1. AFM micrographics of NBR and DLC films deposited on NBR under different SP.

Fig. 2. Visible (514.5 nm) Raman spectra of the DLC films deposited on NBR under different SP. The glossy lines represent the fitting of D and G peaks for the film under 1.8 Pa.

bond disorder and presence of sp^2 rings or chains, can be derived by data of the fitted curve including G-peak position and the area ratio of D peak to G peak (I_D/I_G). The schematic variation of fitted G-peak position and I_D/I_G corresponding to different specimens is shown in Fig. 3. It can be seen that the G-peak position slightly increased from 1540 cm^{-1} to 1560 cm^{-1} approximately, but I_D/I_G decreased from 1.0 to around 0.3 with increasing SP. Accordingly, G-peak position is considered to depend on the bond disorder and the presence of sp^2 rings or chains: the increase of sp^2 chains has a direct influence on increasing G-peak position, whereas the result of bond disorder is the opposite [26]. The information of G-peak position (in Fig. 3a) indicates that the number of sp^2 chains grows with the increasing SP, although the effect on G-peak position of bond disorder is also nonnegligible. Normally, the I_D/I_G

should be < 0.2 in the Raman spectra with incident lights at wavelengths of 514.5 nm if the fraction of sp^3 bonds exceeded 20% [26]. It is considered that the amorphization trajectories of as-synthesized DLC films belong to the stage between nanocrystalline graphite to a-C, and the structure of them consists of mainly sp^2 bonds in puckered ring-like configurations (consisting of five-, six-, seven-, and eightfold disordered rings), and few if any sp^3 bonds [27–30]. Combining with the trend of I_D/I_G (in Fig. 3b), it can be referred that presence of sp^2 rings reduces and the number of sp^3 bonds increases as SP rises [31]. Accordingly, it is true that, in both hydrogen-free and hydrogenated DLC deposition processes, the sp^3 bonds are more efficiently formed at relatively high kinetic energies of incident species. Furthermore, the sp^3 bonds are formed predominantly in the subsurface region and the sp^3 formation process in either hydrogen-free or hydrogenated DLC deposition may be referred as a sub-plantation process [32]. However, excessive energy of incident species in the MS process also impedes the formation of sp^3 bonds. In this case, a high SP promotes the formation of sp^3 , because it tailors the energy of the carbon atoms to a suitable value.

The FTIR spectrum is used to identify the functional group and present different vibrational modes of various bonds presented in the films. Fig. 4 shows the FTIR spectrum of NBR substrate and as-synthesized DLC films on NBR. A medium band at 1241 cm^{-1} (assigned to $sp^3\text{ C-C}$) appears in the spectra of all DLC films, indicating the existence of tetrahedral carbon structures in the DLC films. A peak at 1650 cm^{-1} is assigned to the stretching vibration of $\text{C}=\text{C}$. As for NBR whose band at 1650 cm^{-1} is stronger than that in DLC films, the primary reason is that NBR consists of cis-butadiene, which contains non-central symmetrical $\text{C}=\text{C}$. It is important that the peak intensity reflected by $\text{C}=\text{C}$ is gradually weakening as the SP increases in the DLC films, in compatible with the results of the Raman spectra (Fig. 2). In addition, the weak peak at $\sim 723\text{ cm}^{-1}$ belongs to the out of plane bending configuration of the graphite-like carbon [33], which only appears among DLC specimens rather than NBR (seen in the red rectangular box in Fig. 4).

Fig. 5 compared the water CA of NBR and DLC-coated NBR. The result shows that both NBR and DLC films exhibit hydrophobic properties. In all cases, the CA is larger than 105° , and an increase in the

Fig. 3. (a) G-peak position and (b) I_D/I_G ratio of the fitted Raman spectra of all DLC films. The continuous curves are obtained by polynomial fitting, in order to reflect the changing trend.

Fig. 4. FTIR spectra of the NBR and DLC films. A local enlargement of the absorbance from 700 to 750 cm^{-1} is inserted in the red rectangular box. And all spectra were shifted vertically in order to differentiate them clearly. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

water CA was evident after NBR coated with DLC. Particularly, when SP is 0.2 Pa, CA reaches the maximum value of about 118° in all samples. The improved hydrophobic performance by DLC films can effectively prevent contamination from sewage in most applications. The dispersed holes observed in the DLC surfaces (Fig. 1) can influence the final value of the CA measured, which is reflected in the standard deviation of the samples. On the other hand, DLC films prepared under different SP present different surface wettability. A higher surface roughness usually leads to a lower CA. Therefore, the differences in surface wettability of DLC films obtained under various SP were affected by surface roughness.

The total surface energy and its corresponding components for surface of NBR and DLC-coated NBR were calculated by using a theoretical method (OWRK [23]). In the OWRK method, demineralised water (polar) and diiodomethane (non-polar) were only used in order to minimize the effects of uncertain surfaces (surfaces with a low surface energy, polar or non-polar surfaces) [34]. Corresponding results are shown in Fig. 6. The results show that all the surfaces can be attributed as a low-energy surface as their surface energy is $< 25 \text{ mJ/m}^2$. When comparing all DLC-coated NBR with uncoated NBR, we can see that the

Fig. 5. Steady-state values of contact angles on NBR and DLC films under different SP, wetted with water.

Fig. 6. Total and component surface energies of NBR and DLC surfaces deposited under different SP, calculated by the OWRK method.

influence of SP on surface energy is remarkable. As the SP increases, the total surface energies first increase to a maximum when SP is 1.4 Pa and then decrease. The roughness will increase if the surface energy is high

because the atoms need the high energy to agglomerate and form the film. Therefore, the final roughness of the DLC films is strongly dependent on the surface energy [35]. The result agrees well with the previous AFM results. It is worth mentioning that the added surface energy of solid can enhance the wettability of the liquid on solid surface, which facilitates the application of DLC films in liquid lubrication. For example, some oil-DLC surfaces show preferable characteristics in tribology [36,37]. Fortunately, the appropriate increment of surface energy still maintained the hydrophobic properties of DLC films (Fig. 5).

Fig. 6 also shows that the dispersive components of the surface energy are rather close to the total surface energy among all samples. There are almost no polar components of surface energy on DLC surfaces. Therefore, the DLC's differences in terms of the surface energy are mainly due to the dispersive component of the surface energy, not the polar or total component. This indicates that the dispersive component tends to play the key role in tailoring the performance of the DLC films where the wettability (solid-liquid interactions) is concerned.

Generally, the adhesion strength of a DLC film is evaluated in terms of a critical load at which an instant failure between the film and the substrate occurs [19,38]. However, for a rubber substrate the critical load cannot be well defined because of rubber's low rigidity. As shown in Fig. 7, the scratches on DLC-coated NBR were investigated. It can be seen that there was a distinct delamination (marked by red elliptical circles) at the end of all the scratches (larger load), which was caused by the large strain of NBR. However, in the initial stage of scratches (small load), it is different. For instance, in the small load areas, comparable delamination occurred in P10 and P18, but for P14, there were just some minor cracks, which indicated good adhesion of P14. Besides, the status of the other two specimens was between above-mentioned situations. It can be deduced that the increased adhesion strength of the DLC films impede the possible delamination of the films in the scratch

Fig. 8. Tribological properties of the uncoated and DLC-coated NBR.

process.

The evolution of the coefficient of friction (CoF) as a function of the number of revolutions during the tribo-testing of uncoated and DLC-coated NBR is shown in Fig. 8. First of all, it can be stated that all applied coatings resulted in decreased values of the CoF (under 0.4 during the whole test) of the modified NBR substrates. Compared to uncoated NBR, DLC-coated NBR shows less fluctuation in the CoF during its evolution due to the declined roughness. It is noted that there is a reduction of average CoF in DLC-coated NBR with the increase of SP, but the exception is that P14 and P18 are comparable. The overall contribution to CoF is due to the adhesive interaction and the hysteresis

Fig. 7. SEM micrographs showing surface morphologies of scratches on DLC-coated NBR. The red elliptical circles indicate where typical delamination of the films appears. The load in the micro-scratch test increases gradually from left to right in each diagram. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

Fig. 9. Wear profile of the wear tracks for the uncoated and DLC-coated NBR. The labels R and M define the range of wear tracks.

contribution [39]. Roughness of the film seems to have a positive influence on its frictional characteristics due to the reduction of friction in the contact area during the tribo-testing [40], which in turn reduces the adhesive contribution to friction, resulting in a stable CoF lower than 0.25 of P14. Moreover, as the sliding time increases, the CoF of P10 and P18 tended to increase in the latter stage, not as stable as other coated samples. This phenomenon may be attributed to the difference in adhesion. The delamination of DLC films will cause the increase the CoF, therefore excellent adhesion can ensure that films maintain stable performance in a sustained wear condition.

Fig. 9 shows the profile of the wear tracks after tribo-testing. Particularly, the uncoated NBR that has lost the most material as confirmed by the highest wear depth and also by the highest CoF, which is indicative of poor wear resistance. In DLC-coated NBR, the depth of wear is too shallow to distinguish each other as compared with roughness. In order to discuss the triboperformance of the various DLC films, the laser confocal microscopic images under different amplification multiples for all DLC-coated NBR are compared in Fig. 10. For all samples, the images provide good visualization of the microstructures of worn and unworn regions on their surfaces. It can be seen from Fig. 9 that there are many cracks on the surfaces of samples probably due to the expansion of NBR in the sputtering process. Inside the wear track, the patches reduced in size due to brittle fracture happening within the patch-like DLC film during the tribo-testing. Closer observations illustrated that the density of cracks obviously decreases when SP increases to 1.4 Pa, which is attributed to the high proportion of sp^3 bonds in sample P14 and P18. It is also observed that the worn part exhibited white under the microscope, and the phenomenon always occurred around cracks. The slight whitish wear area confirms the good wear resistance of the P14 and P18

during the tribo-testing under the given load, compared to the others.

The surface roughness was reported to reduce the CoF by suppressing wear particle generation, removing the wear particles from the sliding interface and preventing the agglomeration of wear particles. The reduced contact area between the sliding ball and the rough target surface also decreased the CoF and wear rate [41]. On the other hand, it has been reported in literatures [42,43] that the DLC transfer layer acts as a solid lubricant at the tribo-contact, thereby yielding good frictional performance during the tribo-testing. Such formation of a transfer layer is attributed to the high total surface energy (seen in Fig. 6), which potentially favoured an adhesive interaction during the tribo-testing.

4. Conclusions

DLC films with enhanced friction performance were successfully deposited on NBR by using the DC-MS. The effects of the SP on the surface topography, structure, wettability and tribology characteristics of the DLC films were investigated in details. The DLC film prepared under 1.4 Pa exhibits enhanced triboperformance leading to a stable CoF < 0.25, which is very promising for practical tribological applications. The observed improved friction performance of DLC sample prepared under 1.4 Pa is ascribed to its low CoF and high wear resistance, which attributes to its high proportion of sp^3 bonds and surface energy. An increasing SP contributes to the tribological properties of DLC films on NBR, but an excessively high SP instead hinders the formation of films. Therefore, an optimal SP is crucial in MS.

Fig. 10. Screen shots of images (magnification(s): 10×) of the wear tracks on coated-NBR. Images (magnification(s): 20×) are shown on the right side of corresponding images.

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