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Rapid Note

Adhesion and its influence on micro-hardness of DLC and SiC films

Q.R. Hou[1,a], J. Gao[1], and S.J. Li[2]

[1] Department of Physics, The University of Hong Kong, Pokfulam Road, Hong Kong
[2] Department of Materials Science and Engineering, Beijing University of Aeronautics and Astronautics, Beijing, P.R. China

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Abstract. Micro-hardness and scratch adhesion testing are the most commonly used techniques for assessing the mechanical properties of thin films. Both of these testing methods utilize single-point contact and induce plastic deformation in the substrate and film. However, the influence of adhesion on the measured hardness has been seldom reported so far. In our experiments, diamond-like carbon (DLC) and silicon carbide (SiC) films deposited on silicon and nickel-based alloy substrates by pulsed laser ablation were indented and scratched by a Vickers micro-hardness tester and a diamond-cutter, respectively. It was found that the composite hardness decreased more rapidly for poor adhesion when increasing the indentation load. The result was explained by the elastic-plastic deformation mode of indentation and helped us to understand the physical meaning of one parameter commonly introduced in the models used to separate film hardness from the composite hardness.

PACS. 62.20.Qp Tribology and hardness – 68.35.Gy Mechanical and acoustical properties; adhesion – 81.15.Fg Laser deposition

Adhesion and hardness are the two important mechanical characteristics for various coatings. Micro-hardness and scratch adhesion testing are the most commonly used techniques for assessing these properties of thin coatings. Due to the small thickness, however, the measured hardness is usually influenced by a number of factors as well as adhesion [1]. The most important ones of these factors include film thickness, indentation depth, substrate and film hardness, *etc.* So far, several models have been proposed to separate the film hardness from the composite hardness [2–5]. Jonsson and Hogmark used an area law of mixtures while Burnett and Rickerby developed a volume law of mixtures. He *et al.* proposed a model based on the function of depth weight factor and combined the above two models. According to the model reported by He *et al.*, the composite hardness is the following

$$H_c = H_s + [(m+1)t/mbD - t^{m+1}/mb^{m+1}D^{m+1}](H_f - H_s), \quad (1)$$

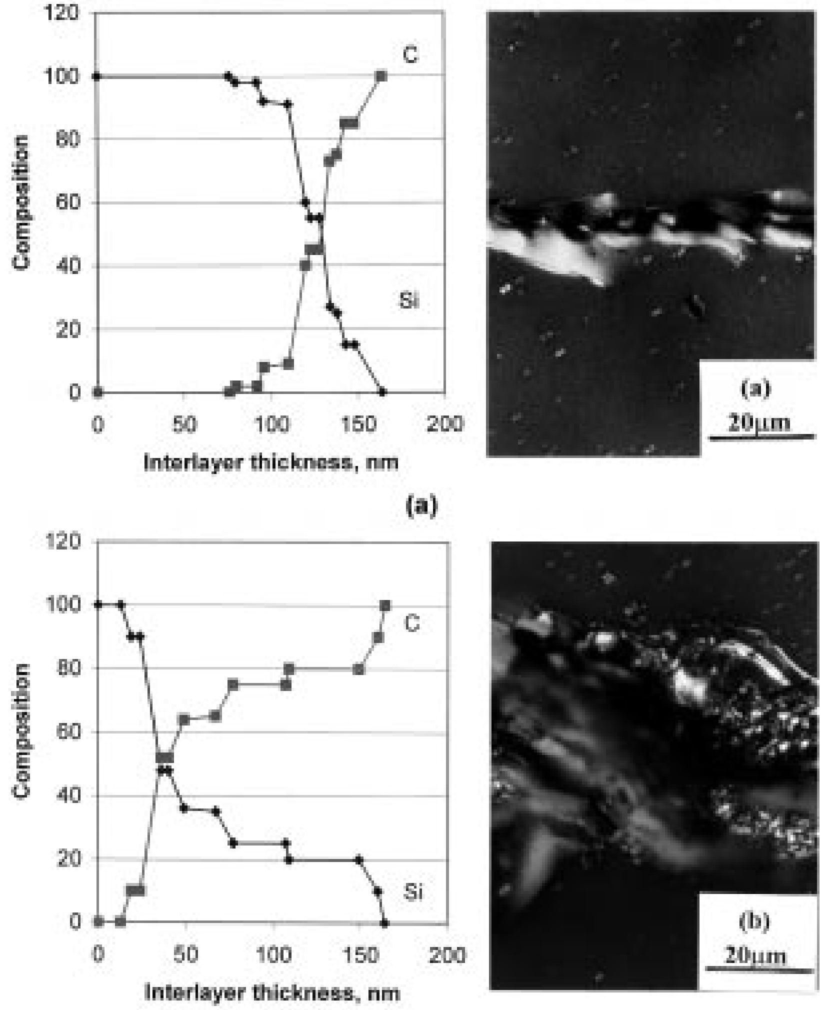
where H_s and H_f are hardnesses of the substrate and the film, respectively, t is the film thickness, D is the indentation depth, m is the power index, and b is the critical reduced depth beyond which the material will have no effect on the measured hardness. Equation (1) is exactly the same as the developed area law of mixtures when m is set as 1 and the volume law of mixtures while m is set as 2 [2]. He *et al.* argued that the influence on hardness must be related to both load supporting area and plastic deforming volume and found that m was actually 1.2 for hard films on soft substrates. We noticed that b has different values even for the same film-substrate combinations such as diamond-like carbon (DLC) films on silicon substrates as shown in Table 1.

According to the elastic-plastic deformation mode of indentation, the plastic zone morphology beneath indentation is typically hemispherical and extends to some distance beyond the edge of the indentation [1]. The critical reduced depth b (the ratio of the plastic zone radius to the indentation depth) is determined by the ratio E/H of Young's modulus to hardness, thus

$$b \propto (E/H)^n \quad (2)$$

where n is between around 0.33 and 0.5 [3]. It is easy to understand why b has different values for different film/substrate combinations. The reason for different values of b for the same film/substrate combination is not clear at this moment. In this work, we investigated the influence of adhesion on the values of b for the DLC/Si and SiC/Ni-based alloy systems. For each system, two samples

[a] e-mail: qhou@hkusua.hku.hk



(b)

Fig. 1. The correlation between the composition change of the intermediate layer (left side) and the scratch adhesion of the DLC film (right side). The adhesion is good in case (a) and poor in case (b). The vertical load during the scratch was 5.5 N.

were prepared with different adhesion by changing the interface conditions.

Diamond-like carbon and silicon carbide films were deposited on silicon and Ni-based alloy (GH-128) substrates by pulsed laser ablation of graphite and SiC targets, respectively [6,7]. The background pressure of the chamber was below 3×10^{-6} mbar. The distance between the substrate and the target was typically 5 cm. An excimer laser with wavelength of 308 nm was used. The laser pulse energy and repetition rate were 230 mJ and 5 Hz, respectively. The laser beam was focused by a lens onto the target at a density of 4 J/cm^2. The thickness of the film was measured by a Dektak^{3}ST surface step profiler. The micro-hardness measurements were carried out on a Shimadzu hardness tester (Vickers). On each specimen indentations were made with five loads ranging from 25 g to 300 g and at least four impressions were made at each load. According to equation (1), the critical reduced depth b was readily determined when other parameters were known or measured. In the experiments, H_c and H_s were measured by the hardness tester (Vickers), t was measured by the surface step profiler, the diagonal d of the impression was measured by the optical microscopy of the hardness tester and $D \approx d/7$, $m = 1.2$ [2], and H_f was measured on thick films ($t > 1$ μm) under a load of 25 g by the same hardness tester. For DLC films ($t = 1145$ nm, 2580 nm), the film hardness H_f was about 38 GPa [9]. For SiC films ($t = 2480$ nm, 2200 nm), the film hardness H_f was about 25 GPa which was very close to the results reported elsewhere [10]. Adhesion of the films was studied by a simple scratch method [5]. In our approach, a diamond-cutter was drawn across the film under a fixed vertical load. The width of the scratch track measured by optical microscopy and the surface step profiler indicated adhesion of the films. A good adhesion corresponded to a narrow scratch track. Otherwise the width was broad since the film was gouged out along the scratch track border during the scratch. Adhesion of the films can be enhanced by introducing a composition-graded intermediate layer. The parameters of these films and interfaces are listed in Table 2.

For DLC film deposited on silicon substrate, adhesion can be controlled by changing the composition gradient in the intermediate layer as shown in Figure 1 [8].

Table 1. Values of the critical reduced depth b for different film/substrate combinations.

Reduced depth b	1	5.6	7.1	13.7
Film/substrate	DLC/Si	DLC/Si	Cr/steel	Cr/Cu
Deposition method	Laser ablation	Sputtering	Sputtering	Sputtering
Reference	[5]	[2]	[4]	[4]

Table 2. Parameters of the grown films and intermediate layers.

Film/substrate	DLC/Si	DLC/Si	SiC/Ni-alloy	SiC/Ni-alloy
Film thickness (nm)	753	591	755	590
Interlayer thickness (nm)	164	164	29	0
Adhesion	Good	Poor	Good	Poor
Reduced depth b	5.4	4.0	9.7	4.8

Figure 1a shows that the film has a good adhesion while Figure 1b indicates poor adhesion of the film. The corresponding micro-hardness difference $\Delta H = H_c - H_s$ is plotted against the ratio t/d of film thickness to diagonal of the indentation ($d = 7D$) in Figure 2. The critical reduced depth b was about 5.4 for good adhesion and 4.0 for poor adhesion. It is clearly seen that the micro-hardness difference decreases more rapidly for poor adhesion when increasing the indentation load (*i.e.* small value of t/d). Besides, it is difficult to measure the micro-hardness of the film with a poor adhesion at large indentation loads due to the easy delamination of the film.

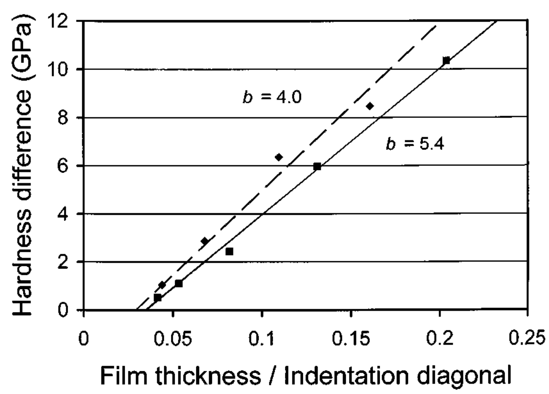


Fig. 2. The micro-hardness difference $\Delta H = H_c - H_s$ *versus* the ratio t/d of film thickness to indentation diagonal for DLC/Si combinations. The critical reduced depth b was about 5.4 for good adhesion and 4.0 for poor adhesion.

For combinations of SiC and Ni-based alloy, one sample was prepared by direct deposition of SiC film on the substrate while another one with a composition-graded intermediate layer [7]. Adhesion was greatly enhanced when the film had an intermediate layer as shown in Figure 3a. The width of the scratch track was very narrow. The adhesion of the film without an intermediate layer was very poor as shown in Figure 3b. The width of the scratch track was broad. Under an optical microscope, cracking and buckling of the film were also observed, indicating the large residual stress in the film. No cracking or buckling was found when the intermediate layer was used. The micro-hardness increase $\Delta H = H_c - H_s$ *versus* the ratio t/d is presented in Figure 4. The critical reduced depth b was about 9.7 for good adhesion and 4.8 for poor adhesion. Similar to the behavior observed in Figure 2, the micro-hardness difference decreases more rapidly with increasing the indentation loads for poor adhesion.

As a rule-of-thumb, DLC films prepared by sputtering deposition were more adherent than those made by pulsed laser ablation due to the sputter cleaning effect before film deposition. The reported low value of b by Hou and Gao [5] may be due to the very poor adhesion of DLC films, since the films were obtained by pulsed laser deposition without an intermediate layer.

According to equation (2), if materials with different hardness and modulus are deposited on the substrate and the composite structure is indented, the plastic deformation within the film will not have the same radius as that in the substrate. Figure 5a illustrates this schematically.

The radial strain profile in both grown film and substrate calculated by using the elastic-plastic indentation theory would show a strain discontinuity across the interface [11,12]. While this is acceptable for coatings with no interfacial adhesion, when a rigid adherent interface is present, no strain discontinuity can be tolerated [1]. Consequently, the deformation geometry shown in Figure 5a will be distorted. The softer substrate will be constrained to take up the strain profile of the film at the interface, resulting in a change in the substrate deformation zone morphology and a corresponding large value of b as depicted in Figure 5b.

In summary, adhesion influences the micro-hardness of films for hard films deposited on soft substrates. When increasing the indentation load, the composite hardness decreases more rapidly for poor adhesion. The critical reduced depth b is determined not only by the ratio E/H of Young's modulus to hardness [3], but also by the adhesion

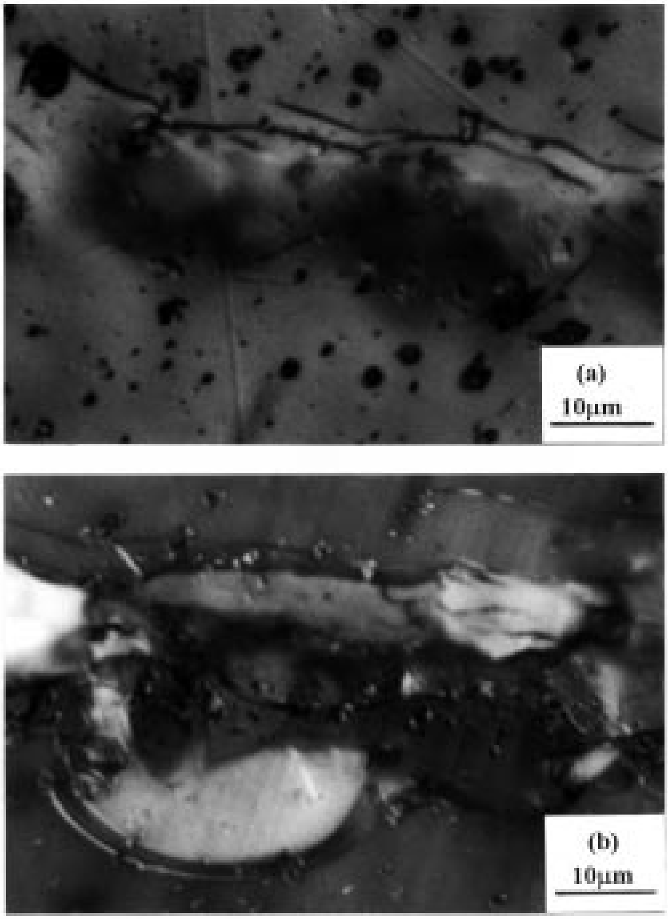


Fig. 3. Optical microscopy images of the scratched SiC films on Ni-based alloy substrates with (a) and without an intermediate layer (b). The adhesion is good in case (a) and poor in case (b). The vertical load during the scratch was 2.5 N.

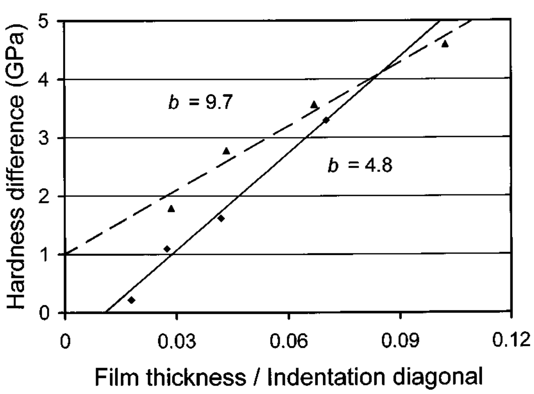


Fig. 4. The micro-hardness difference $\Delta H = H_c - H_s$ *versus* the ratio t/d of film thickness to indentation diagonal for SiC films deposited on Ni-based alloys. The critical reduced depth b was about 9.7 for good adhesion and 4.8 for poor adhesion.

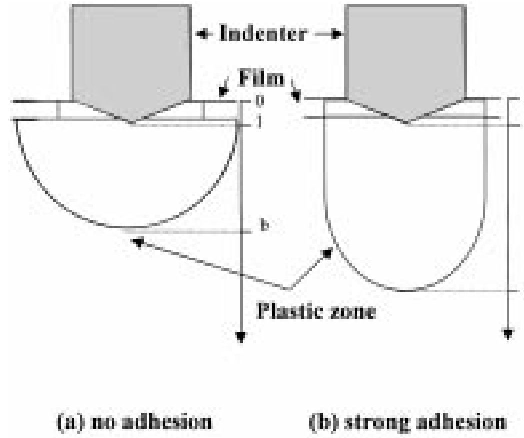


Fig. 5. (a) Schematic representation of deformation associated with indentation in a coated substrate (no adhesion). The critical reduced depth b is governed by the ratio E/H of Young's modulus to hardness as well as adhesion. (b) The effect of a strong film-substrate interface on the determination of morphology in (a).

as well. For the same film/substrate combinations, a large value of b usually corresponds to a good adhesion of the films.

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