Enhancement of impact resistance of CVD diamond coating by multilayer strategy

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Abstract. Poor impact resistance of diamond coating has limited its wide application. In this work, an experimental analysis was conducted to explore the effect of multilayer strategy on impact behaviour of diamond coating. Multilayer diamond coating facilitated the enhancement of impact resistance. Thereafter, four types of chemical vapor deposition coating were deposited to further verify the impact resistance of multilayer diamond coating. Scanning electron microscopy, Raman spectrometer and X-ray diffraction equipment were used to characterize the diamond coatings before and after the impact, respectively. The results showed that MCD/NCD coating had the smallest impact scar. Furthermore, it could be concluded that the multilayer strategy was beneficial for coating to improve the impact resistance.

1. Introduction

Diamond has gained an extensive attraction due to its excellent material properties, such as high hardness, low friction coefficient, and good wear resistance, which makes it be suitable candidate for anti-wear coating [1]. However, the fatal drawback of diamond is fragile and has poor toughness. Hence, for the fragile diamond coating, it is a challenge to avoid the coating failure when the drastic external force, especially impact, is loaded. Impact resistance becomes a big technique barrier for diamond coating regarding its wide-spread utilizations [2, 3].

Many factors affect the impact resistance of coating [4]. Among them, the adhesive strength between coating and substrate partly determines peeling and delamination of coating. The experimental results show that microcrystalline diamond (MCD) coating exhibits the good adhesive strength because the large-sized crystalline grains enhance the so-called mechanical interlock between coating and substrate [5]. However, the coarse surface of MCD coating leads to the high friction coefficient. In order to overcome the drawbacks of MCD coatings, multilayer strategy has been proposed. In this way, multilayer diamond coating presents some superior mechanical properties. In the mechanical engineering industry, especially in machining the difficult-to-machine materials, cutting tools often suffer from the impact. In the mechanical engineering industry, our previously experimental results have found that the insert coated multilayer diamond coating has the better wear resistance and durability than that coated the monolayer one. Obviously, such enhancement on mechanical properties mainly attributes to the multilayer structure. The top-layer coating has two common types: the MCD and the nanocrystalline diamond (NCD). The MCD coating is grown as the top-layer coating to
enhance the mechanical interlock, and the NCD coating is deposited as the top-layer coating to reduce the surface roughness. This is because the NCD coating is conducted to decrease the size of diamond crystalline grain and has the lower friction coefficient than MCD coating. However, nano-sized diamond crystalline grains considerably reduce the mechanical interlock, which weakens the adhesive strength of coating [6].

The purpose of this work is to study the effect of the multilayer and the impact on behaviour of diamond coating. Impact test is performed for multilayer diamond coatings (MCD/MCD and MCD/NCD). For comparison, MCD coating is also tested. The surface morphologies and impact scars are observed by SEM, and the impact resistance is evaluated by an analysis of topographic characteristics of impact scars.

2. Experimental methods

2.1. Fabricating diamond coating

The silicon carbide flat square plane with the 12×12×5 mm was selected as the substrate to deposit diamond coatings which was performed by the conventional hot filament chemical vapor deposition (HFCVD) technique. Prior to diamond disposition, all of the substrates were subjected to the following surface pre-treatments to enhance the diamond nucleation density: (i) polishing by abrasive paper combining with diamond particles (50 μm) slurry for 20 min; (ii) rinsing with deionized water in the ultrasonic vessel for 20 min; (iii) putting the substrate into high-purity acetone with ultrasonic treatment.

The reactant gas was an acetone-H2-Ar gas mixture. Six tantalum wires were used as the hot filaments to offer the enough high temperature for CVD diamond coating growth. All the tantalum wires were dragged straightly by the springs to retain the constant distance from the substrate at the high temperature. During the deposition, the hot filaments maintained about 2200 °C. The distance between hot filaments and substrate was fixed at about 12 cm. In this way, the temperature of substrate surface was about 800-900 °C, which is the suitable temperature range for growing the diamond crystalline grains. All the deposition parameters of CVD diamond coatings were listed in Table 1.

Table 1. Deposition parameters of diamond coatings.

| Item                | MCD          | NCD          |
|---------------------|--------------|--------------|
| Acetone/H2/Ar flow (sccm) | 70/200/0     | 70/200/100   |
| Pressure (Torr)     | 30–37        | 7–15         |
| Bias current (A)    | 3            | 3            |
| Duration (h)        | 4            | 4            |

For the as-deposited diamond coatings, the surface morphologies were evaluated by the Hitachi SP 2600 SEM (Hitachi, Ltd.). The phase of diamond was analysed by Horiba Jobin Yvon Xplore Raman spectroscopy (Horiba Jobin Yvon Ltd.) with an excitation wavelength of 632.8 nm.

2.2. Impact testing

In this work, the impact resistant was also evaluated, and an impulse of force was used to mimic impact. It needed pointed out that this type of impact was different from the impact fatigue test where the various cyclic loads were applied to assess the diamond film’s adhesive strength. In this work, the impact was generated by the bump of a free-falling body. A solid weight was used as the free-falling body (4.43 m/s). A tube remained stationary and was perpendicular to the surface of diamond coating. Then, the weight could fall down along the internal surface of tube to guarantee the vertical bumping on the punching head that touched on the diamond coating at the beginning of testing and also could move smoothly along the vertical direction. After bumping the punching head, an impact was imposed on the diamond coating. In order to obtain the identical impact, the distance (h) between weight and punching head kept the same magnitude.
3. Results and discussion

3.1. Characteristic of multilayer diamond coating

Fig. 1 showed the surface morphologies of the as-deposited diamond coatings. The MCD coating had a very rough surface, as shown in Fig. 1 (a). Many micro-sized diamond grains were grown on the substrate, which enhanced the mechanical interlock between MCD and substrate. As a result, the adhesive strength could be improved. This was also favourable for the impact resistance. For NCD coating, the size of diamond grains considerably decreased to the nano scale. The surface morphology exhibited the cauliflower shape, as shown in Fig. 1 (b). The nano-sized diamond grains were useful to smooth the surface of coating. However, its weak mechanical interlock led to the decrease in adhesive strength, which made a challenge for NCD coating to resist external load. For the MCD/NCD and MCD/NC coating, SEM images of each MCD and NCD layer had the similar surface morphology as the monolayer MCD and NCD coatings, as shown in Fig. 1 (c) and Fig. 1 (d), respectively. However, these sandwich-structured coatings showed a combining advantage of adhesive strength and friction coefficient. The MCD coating was used to improve the adhesive strength due to the strong mechanical interlock, and the top layer of NCD coating could decrease the surface roughness to obtain low friction coefficient. The cross-sectional morphology of MCD and NCD coating was shown in Fig. 1 (e) and Fig. 2 (f) respectively. The thickness of coating was about 10μm. In the cross-sectional morphology of MCD/MCD and MCD/NCD coating, the thickness of each layer, including MCD and NCD coating, was about 5-6μm, as shown in Fig. 1 (g) and Fig. 1 (h), respectively.

![SEM images of as-deposited coatings](image)

Fig. 1. SEM images of as-deposited coatings: the surface morphologies the surfaces and cross-sectional morphologies of (a) (e) MCD, (b) (f) NCD, (c) (g) MCD/MCD and (d) (h) NCD/MCD deposited on silicon nitride substrates, respectively.

The XRD pattern were performed for as-deposited diamond coatings to detect the crystalline structure of diamond coating, as shown in Fig. 2. All the samples had the two diamond peaks at about 44° and 75°, which were assigned to (111) and (220) reflections of diamond.
The quality of diamond coating was determined by Raman spectra. Fig. 3 showed the Raman spectra of all the diamond coatings. The sharp and narrow peak was found at the ~1332 cm$^{-1}$ for the MCD coating, which corresponded to sp3 carbon bond. For all of the NCD coatings, the two obvious peaks are found in their Raman spectra. One was located at the ~1332 cm$^{-1}$, the other was at the ~1580 cm$^{-1}$ that denoted the G carbon bond [7]. In this analysis, the NCD coating was not a pure diamond material. The non-diamond phases may grow in the boundary of diamond grains. Thus, the hardness of NCD coating was less than that of MCD coating. In addition, the total residual stress of each coating could be calculated from the Raman spectra by the shift of the stressed diamond Raman line relative to the natural diamond line at 1332 cm$^{-1}$ [8]. Here, it should be pointed out that although Raman spectra could only offer the information of several micron depth of top layers, it was still a reasonable and convenience method to relatively quantify residual stress for coating. Then the total residual stresses were calculated as -0.767 GPa for the MCD coating, -9.72 GPa for the NCD coating, -1.93 GPa for the MCD/MCD coating, and -7.938 GPa for the MCD/NCD coating. Obviously, the large residual stress was harmful to impact resistance. However, multilayer coating seemed to be beneficial for decreasing the residual stress between the NCD and MCD/NCD coating.

Note that the relative soft NCD coating was favourable for impact resistance, but its low adhesive strength easily caused the peeling of coating. Similarly, MCD coating showed the high adhesive
strength, but it had a high hardness. The coating seemed to be difficult to achieve high toughness and high adhesive strength, which brought an interesting issue to pursue a suitable balance between toughness and adhesive strength to enhance impact resistance.

3.2. Surface morphology of impact scar
After impact testing for each sample, the surface morphology of impact scar was observed to evaluate the impact resistance by assessing the damage intensity of coating. Fig. 4 showed the impact scars and EDX of all the examined diamond coatings. For the MCD coating, it could be seen that the coating peeled off from the substrate, as shown in Fig. 4 (a). As mentioned above, MCD coating was a high purity diamond material, which made it have a superior hardness. However, this led to a low toughness. However, MCD coating had a remarkable adhesive strength due to the strong mechanical interlock. In terms of impact resistance, there was a contradiction between low toughness and strong adhesive strength for MCD coating. In fact, the experimental results indicated that the MCD coating could not endure the impact. The toughness seemed to be more important than adhesive strength for impact resistance.

For the NCD coating, the area of the peeling coating was smaller. Only some cracks appeared in the range of impact scar, as shown in Fig. 4 (b). Obviously, the NCD coating presented a better impact resistance than the MCD coating. Non-pure diamond grains of the NCD coating caused the reduction in the hardness and thus increased the toughness. However, NCD coating had a weak adhesive strength. Thus, NCD coating also struck the contradiction between strong toughness and low adhesive strength. The results demonstrated again that the toughness was of importance in impact resistance of coating. For the bilayer MCD/MCD coating, the impact scar was similar with the MCD coating. In addition, the white areas were formed around the shedding area, which indicated that the top layer of MCD coating was delaminated. Many properties of the top layer of MCD/MCD coating were similar to those of MCD, such as the toughness and adhesive strength. The second layer was less damaged because of the protection of the upper layer. For the MCD/NCD coating, many scratches could be found, but the delamination or cracks did not occur. Some large scraps from punching head scattered around the impact scar, as shown in Fig. 4 (c). The results suggested that the bilayer diamond coating had a great improvement on impact resistance in comparison with the monolayer diamond coating. Based on the results of the impact testing of NCD coating, the top layer of NCD coating played an important role in the improvement of impact resistance of MCD/NCD coating. The good toughness of NCD coating could effectively defend the impact to avoid the serious damage of coating. The integrity without peeling indicated that the bottom layer of MCD coating may not suffer from the more serious impact than monolayer MCD coating. It was possible for the NCD coating to reduce the impact intensity before the impact stress waves reach the MCD coating. Meanwhile, the bottom layer of MCD coating provided the strong adhesive strength to further prevent the coating from peeling off from the substrate. Hence, the MCD/NCD coating presented a perfect impact resistance which was also agreement with our previously experimental observations. As discussed above, the toughness was the main role for impact resistance of coating.

As mentioned above, the enhancement of multilayer coating on impact resistance possibly results from the special mechanical properties. The ranking of impact resistance followed the trend MCD/NCD coating > MCD/MCD coating > NCD coating > MCD coating. The characteristics of the
experiments coincide with our theoretical analysis. Therefore, the conclusion could be drawn that multilayer strategy gave rise to the decrease in composite Young’s modulus. As a result, the toughness of coating was improved, which leaded to the enhancement on impact resistance of the MCD/NCD coating.

Such results were also in agreement with the previous literatures. It was found that the MCD/NCD coated inserts had the longer life time than MCD coated insert during machining Al-Si alloy with high Si content. The indentation tests also proved that the multilayer coating exhibited the enhanced crack resistance and improved adhesive strength as compared to the monolayer coating.

4. Conclusions
In this work, the impact behaviour of multilayer diamond and the novel coating has been studied. Here, the composite Young’s modulus is adopted to characterize the toughness of diamond coating, which is directly associated with the impact resistance. Four different diamond coatings, including MCD, NCD, MCD/MCD and MCD/NCD diamond coatings, are employed as the samples. The calculated data shows that the composite Young’s moduli of MCD, NCD, MCD/MCD, and MCD/NCD coatings are 312, 275, 246, and 222 GPa, respectively. This suggests that the toughness decreases as the number of layers increases. MCD/NCD coating has the better toughness, then it would have the strongest impact resistance.

Bi-layer (MCD/NCD) diamond coating is fabricated as one examined sample. Meanwhile, the mono-layer (MCD and NCD), bi-layer (MCD/MCD) diamond coatings are also deposited for comparison. The results show that the MCD/NCD coating presents a better impact resistance than the mono-layer diamond coating. For the MCD/MCD, peeling is similar to those of MCD coating. For the MCD coating, peeling can be clearly observed. However, for the NCD coating, peeling does not occur, but there are some cracks distributed in the impact scar. The results demonstrate that the multilayer strategy is beneficial for coating to improve the impact resistance, which is in good agreement with our theoretical calculations and the finite element analysis. The results also suggest that multilayer strategy is a potential approach to improve impact resistance of coating, even for the high fragile and hard material.

Acknowledgements
This work is supported by the National Natural Science Foundation of China Grant No. 51405285, Natural Science Foundation of Shanghai Grant No. 18ZR1416100, Capacity Building Projects in Local Universities of Science and Technology Commission of Shanghai Municipality Grant No.1902050900, and Science and Technology Commission of Shanghai Municipality Grant No.19DZ2271100.

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