136 Tribological properties of a-C/a-C:Cr multilayer coatings

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Summary
Alternated a-C/a-C:Cr (about 2.5μm thickness) coatings were made by DC magnetron sputtering from graphite and Cr target in an argon discharge. Mechanical and tribological properties were measured by indentation, scratch and pin-on-disc test. The critical scratch load of a-C/a-C:Cr multilayer coatings for total failure is approach 100 N. The friction coefficient remains within the range of 0.08-0.1 at loads between 10 and 40 N during a pin-on-disc wear test. The wear depth only reaches 0.6 μm after a one hour wear test. The surface of a-C/a-C:Cr multilayer coatings undergoes cyclic deformation, and failure eventually occurs as a result of fatigue. The greater compliance and fracture toughness of the a-C/a-C:Cr multilayer coatings allows greater strains or strain energy to be stored before coating failure, and hence significantly improves wear resistance.

Key words: a-C/a-C:Cr multilayer coatings, wear resistance, magnetron sputtering

1. Introduction
Carbon based coatings, diamond like carbon (DLC) or amorphous hydrogenated (a-C:H) carbon, which show excellent hardness, chemical inertness, low friction and wear resistance, have been successfully used as low load protective coatings for both magnetic and optical discs. These properties also make carbon based coatings promising candidates for the higher load wear protection of machine parts and tools.

A major problem associated with the application of the carbon based coatings is insufficient adhesion on steel. The graded transition from TiN-TiCN to amorphous hydrogenated-carbon has shown some success in eliminating the problems of lattice mismatch and a high stress near the coating/substrate interface(1)(2). To further reduce internal stress and ensure higher performance of carbon based coatings, some studies have advocated metal doping of the carbon layer(3)(4). Dimigen and Klages(5) indicated that Me-DLC films attain a hardness H typical of inorganic ceramics but have Young's modulus value of E significantly lower than that of ceramics. The corresponding H/E ratios are relatively high and their values are typical for polymers. The surface energy of Me-DLC films with low Me contents are also more similar to that for polymers than to those for metals and ceramics. The combinations of high elasticity and H/E with low surface energy could explain the low friction coefficient and low wear rate. Similar conclusion have been conducted by Grill(6). This paper represent the tribological behaviour of a-C/a-C:Cr coating. Failure mechanisms were proposed for these coatings based on the high resolution microstructure examination.

2. Materials and methods
M2 tool steel approximately 25 x 20 mm in area and 2 mm thickness, and Si (100) wafer were used as substrate materials. First, a layer of CrN of about 0.3 μm is deposited, then carbon is gradually introduced to produce a layer of a-C:Cr (about 0.2 μm thickness) by DC magnetron sputtering from a graphite target in an argon discharge. The subsequent a-C layer can be obtained by sputtering from the graphite target. The metal concentration is controlled by varying the sputter power supply. The total film thickness is 2.8 μm and with 10 alternated a-C/a-C:Cr layers.

Elastic modulus and hardness were determined using Nanoindentation tester (MICRO MATERIALS, Nano test 500). Indentation tests were conducted under a depth control mode, i.e. the loading process was reversed at a pre-set depth of 200 nm. A dwell time of 10 seconds was applied to allow for possible time-dependent plastic deformation. The raw data of force-displacement curves was processed to obtain elastic modulus and hardness by using a model proposed by Oliver and Pharr (1992). Microhardness and indentation failure measurements were made using Vickers microhardness tester (Leitz) over the load range 10-500 g. Scratch critical load Lc was carried out using a Teer Scratch Tester ST-2200 with loading rate dL/dt = 100 N/min and the table speed dx/dt = 10 mm /min. Friction and wear measurements were carried out by two different tests: (1) a pin-on-disc machine, using a 5 mm diameter tungsten carbide ball as the pin. The normal load varies from 1N to 80 N, the sliding speed was 250 rpm (the linear speed is 10 ± 2 m/min), and the friction force was monitored using a force transducer. The friction coefficient can be measured from the ratio of friction force to normal load. (2) a reciprocating sliding in multipass mode on a Teer-ST2200 scratch tester using a 5 mm tungsten carbide, and the sliding speed was 150 mm/min. All the
tests were carried out in air at \( \sim 20^\circ \text{C} \), relative humidity was about 35 \( \sim 50\% \). A high resolution Hitachi S-40000 field emission gun (FEG)-SEM was used to examine microstructure and failure mechanisms of various multilayer coatings.

3. Results

Fig. 1 shows a very dense alternated a-C/a-C:Cr multilayer as grown on CrN interlayer. Columnar characters still can be observed in the alternated a-C/a-C:Cr multilayer. X-ray analysis shows that various carbide phases exists in the coatings. The hardness of CrN/alternated a-C/a-C:Cr multilayer coatings is approximately 2050 at the applied load of 25 g using the Vickers microhardness test. The measured hardness and elastic modulus are approximately 6.52 GPa and 97.9 GPa under the maximum applied load of 8.0 mN from the loading-displacement curve. No tensile ring or radial cracks appear in the coatings even at a load of 500 g, as shown in Fig. 2(a). A slight reduction of layer thickness or densification occurs in the alternated a-C/a-C:Cr multilayer as shown in Fig. 2(b). No obvious sliding between columnar a-C/ a-C:Cr boundaries or debonding between growth steps are observed in the coatings. Similar time dependent elastic recovery phenomena have been observed in alternated a-C/ a-C:Cr multilayer coatings.

The critical scratch load of a-C/a-C:Cr multilayer coatings for total failure is higher than 100 N. No severe buckling failure of the coating appears ahead of the indenter even at the end of the load 100 N. Only limited chipping was found along the edge of the scratch track. In the constant load scratch test, only slight shear deformation was observed at an applied load of 30 N (Fig. 3(a)). The tendency of shear deformation becomes more clear with increasing applied load to 50 N (Fig. 3(b)). Further increasing the load to 70 N, interfacial cracking and micro-delamination occur along the a-C/a-C:Cr interface or along the CrN/a-C:Cr interfaces (Fig. 3(c)). No tensile cracks were observed inside or on the edge of the scratch tracks or even in the front of the indenter after a 50 N constant load scratch test (Fig. 4). Microflaking and cracking was observed inside and in front of the coatings when the load was increased to 70 N.

The friction coefficient remains in the order of 0.08-0.1 during the reciprocating multipass tests,

![Fig. 1 Cross-sectional SEM image of a-C/a-C:Cr multilayer coatings, showing dense columnar structure.](image1)

![Fig. 2 Fracture SEM images through indentations on a-C/a-C:Cr multilayer coatings, showing no radial or nest tensile cracks appear on the coating surface even at the applied load of 500 g on hardened M2 substrate.](image2)

![Fig. 3 Fracture SEM images through scratch tracks on a-C/a-C:Cr multilayer coatings after constant load scratch tests, showing shear deformation at applied load of (a) 30 N, (b) 50 N, and (c) 70 N.](image3)
Fig. 4 Fracture SEM images through scratch tracks on a-C/a-C:Cr multilayer coatings after constant load scratch tests, showing no cracks appear along the edge or inside the scratch tracks at applied load of 50 N.

Fig. 5 Fracture SEM images showing the microstructure response of a-C/a-C:Cr multilayer coatings after 50 N x 10 cycles reciprocating multipass test; using 200 μm tip radius diamond as a slider.

Fig. 6 (a) The microstructural responses of a-C/a-C:Cr multilayer coatings after 50 N x 520 cycles repeated traversals, (b) near the edge, (c) near the center of wear tracks: using 200 μm tip radius diamond as a slider.

Fig. 7 High resolution SEM images showing the worn surface of a-C/a-C:Cr multilayer coatings after 10N x 60 min pin-on-disk wear test; using 5 mm dia. WC/Co ball as a slider.

using a 200 μm tip radius diamond as the indenter. No obvious failure occurs even after 30 N x 100 cycles of the wear test. Only limited microflaking and chipping was observed on the worn surface after 30 N x 520 cycles sliding test. The layered columnar structure still remains at the centre of the wear tracks. Under an extremely high contact pressure (50 N), only very limited chips were observed along the edge of the wear track after a 10 cycle test (Fig. 5). After 520 cycles sliding test, obvious substrate extrusion from both side of the wear track was observed (Fig. 6(a)). The phenomena of compaction and extensive plastic flow of alternated a-C/a-C:Cr multilayer coatings can be seen to accommodate substrate deformation (Fig. 6(b)).
Microflaking and microbuckling have also been observed on the worn surface. The appearance of a columnar and layered structure disappears near the centre of the wear tracks after 50 N x 520 cycles traverses, as shown in Fig. 6(c).

Fig. 8 High resolution SEM images showing the worn surface of a-C/a-C:Cr multilayer coatings after 40N x 60 min pin-on-disk wear test; using 5 mm dia. WC/Co ball as a slider

The friction coefficient remains within the range of 0.08-0.1 at loads between 10 and 40 N, using a 5 mm diameter tungsten carbide ball as the pin. Fig. 7 shows that the wear depth was only confined to the contact asperities at a load of 10 N, even after a 60 min wear test. Increasing the load to 40 N, the wear depth only reaches 0.6 μm after a one hour wear test (Fig. 8). Very smooth worn surfaces and limited microflaking can be observed after the wear test.

4. Discussion

An amorphous structure of alternated a-C/a-C:Cr multilayers may have the same function for energy absorption and stress dispersion as has been discussed in the a-C:H layer. This avoids both circumferential and radial cracks and increases the loading capacity.

The columnar alternated a-C/a-C:Cr multilayers accommodate shear and normal forces by viscoelastic, plastic deformation and densification. Columnar grain boundary sliding between adjacent columns and localised decohesion between a-C/a-C:Cr interfaces may allow further deformation and energy dissipation. The energy dissipated by a-C/a-C:Cr multilayer coatings may significantly disperse the radial tensile stress developed at the sides of the indenter and hence avoid tensile cracks along the edge of the scratch tracks. Energy dissipation by the coatings also delays plastic deformation and pile up of the substrate materials and avoids premature partial ring crack development ahead of the indenter.

Excellent wear resistance was obtained with alternated a-C/a-C:Cr multilayer coatings. The viscoelastic and shear deformation behaviour, lubrication and transfer mechanism of a-C/a-C:Cr multilayer coatings may be similar to that which occurred in a-C:H coatings. The surface of a-C/a-C:Cr multilayer coatings undergoes cyclic deformation, and failure eventually occurs as a result of fatigue.

5. Conclusion

The columnar alternated a-C/a-C:Cr multilayers accommodate shear and normal forces by viscoelastic, plastic deformation and densification. Columnar grain boundary sliding between adjacent columns and localised decohesion between a-C/a-C:Cr interfaces may allow further deformation and energy dissipation. The greater compliance and fracture toughness of the a-C/a-C:Cr multilayer coatings allows greater strains or strain energy to be stored before coating failure, and hence significantly improves wear resistance.

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References;