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Adhesion Strength of HFCVD Diamond Coating on WC Substrate Seeded With Diamond and Different Ratios of SiC Powders

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Abstract. The effect of seeding using different mixtures of diamond and silicon carbide (SiC) powders on the adhesion strength of hot-filament chemical vapor deposition (HFCVD) diamond coating on WC-6% Co substrates was studied. Diamond powders with the average grain size of 0.5 µm mixed with various concentrations of SiC powder of 175µm average grain size were employed. Diamond layers were deposited using a production unit of HFCVD technique. The diamond film morphology and the diamond quality were examined using field emission scanning electron microscope (FESEM), X-ray diffraction (XRD) and Raman spectrometer respectively. It was found that the diamond morphologies produced and the qualities were almost the same for all the seeding mixtures of pretreatment. With the blasting technique employed to determine the adhesion strength it was found that the highest diamond film adhesion strength was achieved from the seeding of a mixture of diamond with 5g/l SiC powder.

Introduction

Diamond coated tungsten carbide (WC) tools are considered as ideal cutting tools for their excellent characteristics such as high hardness and wear resistance, low thermal expansion coefficient, low friction and good thermal conductivity [1]. These properties are suitable for high efficiency and precision machining of non-ferrous metals and alloys as well as non metallic materials such as ceramics and graphite. Thin diamond films can be directly deposited onto the WC tools with simple and complex shape with chemical vapor deposition (CVD) technique.

As CVD diamond coatings applied to WC-Co cutting tools, the main challenges are to obtain high adhesion strength and high quality of diamond film. This is due to the strong catalytic effect of Co, which promotes the formation of a graphitic intermediate layer on the substrate resulting poor adhesion strength with the diamond film [2]. At present the most common, easiest and effective method employed to increase the adhesive strength and diamond quality is two-step ultrasonically chemical pretreatment method followed by seeding process to increase further the diamond nucleation density. The substrates are first etched in Murakami solution (10g $K_3[Fe(CN)_6]$ + 10g KOH + 100ml H_2O) to roughen the surfaces followed with a second step etching using hydrogen peroxide acid solution (3ml H_2SO_4 + 88ml H_2O_2) to remove substrate surface Co [3-4]. The beneficial effect of a rough WC–Co substrate surface on the nucleation density and the adhesion of the diamond film have been reported by Haubner et al. [5]. Seeding which is the ultrasonic pretreatment with diamond particle suspensions not only produces more homogeneous morphology but also significantly increases diamond surface nucleation density [6-7].

Further research had shown that by using mixed diamond suspensions during seeding, there is a significant effect when small size diamond powders are mixed together with other particles with a larger size. Y. Avigal [8] has proved that by mixing suspension of diamond powders with larger grain size particles of SiC, B_4C and SiO_2 have greatly improved the diamond nucleation density as compared to just using diamond powders alone. The same result was also obtained when diamond powders were mixed with TaC powders [9] and Ti powders [10-11]. All of these results were produced on Si as the substrate material. Previous works show that combining substrate pretreatment methods together with diamond seeding process is essential to improve the diamond nucleation density. However, hardly found work has been done on applying mixed diamond suspension during seeding on WC substrates to determine its effect on the mechanical properties mainly the adhesion strength between WC substrate and the diamond film. Therefore, in this paper we report the effect of mixed seeding of diamond powders and SiC particles on the adhesion strength of HFCVD diamond layer on WC-6% Co substrates.

Material and Procedure

The samples used in this study were commercial grade WC-6% Co with the grain size range of 1-3µm, disk in shape with 12mm diameter and 4mm thickness. All the samples were cleaned with steam blast and dried with air blast to remove loose residues from the substrate surfaces. The samples were then chemically etched ultrasonically with Murakami solution (10 g $K_3[Fe(CN)_6]$ + 10 g KOH + 100 ml H_2O) for 20 minutes at room temperature to roughen the substrate surfaces followed by steam blasting and then dried with air blast. Subsequently, the surface Co was removed by acid etching (3 ml 96 wt.% H_2SO_4 + 88 ml 40% m/v H_2O_2) for 20 seconds followed by steam and air blasting.

Prior to diamond deposition, the etched samples were seeded ultrasonically for 20 minutes in Tickopur and distilled water (10% concentration) mixed with diamond powders (5µm, 0.8g/l) and silicon carbide (SiC) powders (175µm grain size). The concentrations of SiC were varied at 0, 1, 5 and 10g/l. All the seeded samples were then immediately blasted using steam and dried air. The diamond films were deposited onto WC samples using a production unit of HFCVD under the fixed parameters: pressure (10.2mbar), filament temperature (2200^{O}C), gas CH_4/H_2 ratio (1%), deposition time (30 hours) and current (144A).

All the coated samples were analyzed using FESEM, XRD and Micro-Raman Spectrometer to determine the diamond grain size, morphology and the diamond quality respectively. The blasting tests were also conducted on diamond coated samples to determine the adhesion strength of diamond layer. The adhesion strength of the samples was determined by the time of coating failure as well as the diameter of the flake-off area of the diamond layer.

Results and Discussion

Figure 1 shows the FESEM micrographs of diamond morphology of coated WC samples after seeding at different diamond-SiC powder concentrations. All the samples show very rough surfaces, continuous and well faceted diamond films. However, the diamond grain size changed considerably with the concentrations of SiC powder in the seeding diamond suspensions. The diamond grain size of substrates seeded without SiC powder is much smaller (1-3µm) and more uniform as compared to the diamond grain size of substrates seeded with mixtures of diamond and SiC powder with the range of 1-3µm (Figure 1a). It is believed that diamond powder alone has higher access rate of penetrating into the crevices of WC substrate surface creating more nucleation sites and hence increase the diamond nucleation density which translates into smaller diamond grain size. Meanwhile the diamond grain size seeded with mixtures of diamond and SiC powder is much larger (Figure 1b–d) ranging from 1–5µm. This bigger range of diamond grain size is caused by lower nucleation rate whereby the competition that exists between diamond and SiC powder during seeding permits reduced chances of diamond powder to access the substrate surface, resulting in lower nucleation density and bigger diamond grain size.

XRD analysis shown in Figure 2a indicates that all the diamond films produced from different mixtures of seeding comprise of mainly (111) and (220) morphology. It is clearly shown that, the diamond morphology formed on the WC substrates depends on the HFCVD coating parameters, regardless of the seeding conditions. Meanwhile Figure 2b shows the Raman spectrums which reveal the quality of the coated diamonds. All the spectrums show intense sharp peak at approximately 1335cm^{-1} which is the characteristic of high-phase purity of polycrystalline diamond. The spectrums also show the broad band components at approximately 1510cm^{-1} which is attributed to amorphous carbon with high concentration of sp^2 bonds [12-13]. Figure 2b(i) clearly shows that, seeding process done without SiC powder has produced highest concentration of amorphous carbon with the value of counts are about 3300 counts, whereas for seeding process with the mixture of diamond and SiC powder has lower counts of amorphous carbon. All three different mixtures of diamond and SiC powder show counts below 3000 with the minimum counts of about 1600 obtained from the seeding process with diamond powder mixed with 5 g/l of SiC powder shown in Figure 2b(iii). The presence of SiC powder in diamond seeding reduces significantly the concentration of amorphous carbon. This condition will definitely be one of the factors that increased the adhesion strength.

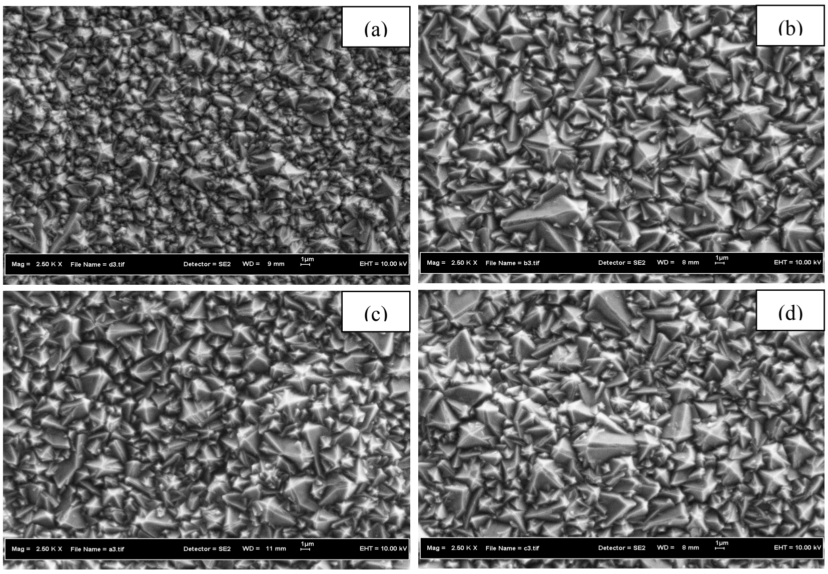
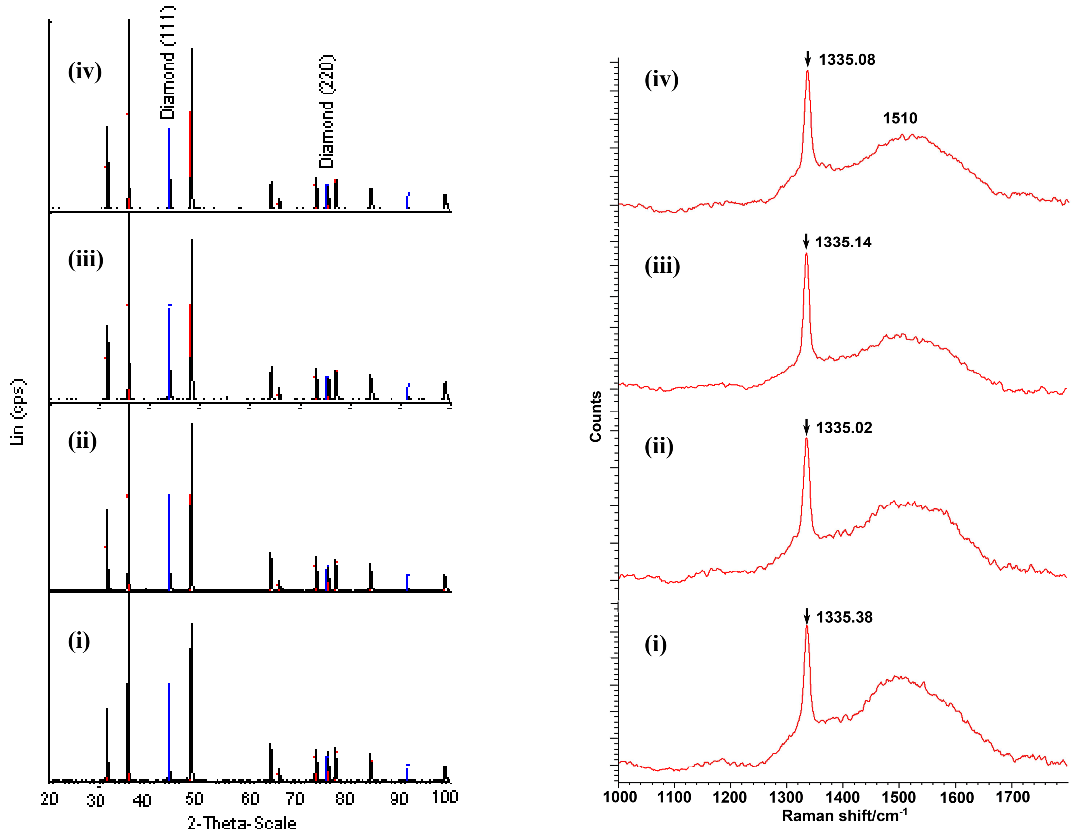


Fig. 1: FESEM images of diamond morphology seeded with 0.8g/l diamond powder and various concentration of SiC particles, (a) 0g/l, (b) 1g/l (c) 5g/l, and (d) 10g/l

Figure 3 shows the results of the blasting tests conducted on the diamond coated samples. Clearly from both Figure 3a and 3b, the maximum time (120s) for diamond film to flake-off and the smallest flake-off size (227µm in diameter) was obtained from the WC sample seeded with diamond and 5g/l of SiC powder. This particular mixture of diamond and SiC powder exhibits the highest adhesion strength was most probably due to the heterogeneous effect or the hammering effect [8], in which the larger and heavier SiC powder impact on the smaller diamond powders at the rough surface of the WC substrate during seeding. This will strongly embed diamond powders into the crevices of the rough surface of the substrate and as a result produce higher mechanical interlocking between diamond film and the WC substrate. The lowest adhesion strength of diamond film seeded with higher concentration of SiC powder (10g/l) was due to the competition between SiC and diamond powder during seeding. With the increasing number and much bigger size of SiC powder making diamond powder more difficult to seed reducing the diamond nucleation rate and also resulting in lower mechanical interlocking which reduced the adhesion strength. Clearly, with no hammering effect of SiC powder for the sample seeded with only diamond powder the adhesion strength was also lower similar to samples seeded with diamond and 10g/l of SiC powder.

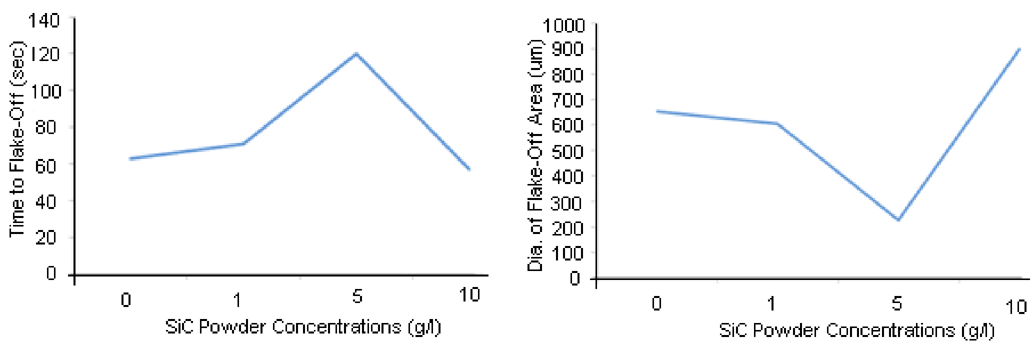
Conclusions

The HFCVD diamond films coated on WC substrates seeded with different SiC particles concentrations mixed with diamond powder produced larger grain size as compared to seeding with just diamond powder. The morphology and quality of diamond films were almost the same for all the samples. However, diamond film seeded with diamond and 5g/l of SiC powder contained less concentration of amorphous carbon as compared to other samples. This is one of the factors that increased the adhesion strength of the sample. From the blasting tests, it is revealed that the diamond film seeded with diamond and 5g/l of SiC powder has the highest strength. It was proven by the longest time to flake-off the diamond film and the smallest size of the diamond flake-off size. In this particular study, it can be concluded that, seeding with a mixture of diamond and 5g/l of SiC powder produces high quality diamond film with the highest adhesion strength. With larger diamond grain size produced, it is very suitable for abrasive applications such as grinding.



(a) (b)

Fig. 2: (a) XRD spectrums (b) Raman spectrums of diamond films seeded with diamond and, (i) 0g/l SiC, (ii) 1g/l SiC, (iii) 5g/l Sic and (iv) 10g/l SiC powder



(a) (b)

Fig. 3: Effect of SiC powder concentrations on, (a) time to flake-off, (b) diameter of flake-off area

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