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## Diamond/ $\beta$ -SiC film as adhesion-enhanced interlayer for top diamond coatings on cemented tungsten carbide substrate

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### ABSTRACT

In present work, diamond/ $\beta$ -SiC composite interlayers were deposited on cemented tungsten carbide (WC-6%Co) substrates by microwave plasma enhanced chemical vapor deposition (MPCVD) using H<sub>2</sub>, CH<sub>4</sub> and tetramethylsilane (TMS) gas mixtures. The microstructure, chemical bonding, element distribution and crystalline quality of the composite interlayers were systematically characterized by means of field-emission scanning electron microscopy (FE-SEM), X-ray diffraction (XRD), X-ray photoelectron spectrometer (XPS), electron probe microanalysis (EPMA), Raman spectroscopy and transmission electron microscopy (TEM). The influences of varying TMS flow rates on the diamond/ $\beta$ -SiC composite interlayers were investigated. Through changing the TMS flow rates in the reaction gas, the volume fraction of  $\beta$ -SiC in the composite interlayers were tunable in the range of 12.0%–68.1%. XPS and EPMA analysis reveal that the composite interlayers are composed of C, Si element with little cobalt distribution. The better crystallinity of the diamond in the composite is characterized based on the Raman spectroscopy, which are helpful to deposit top diamond coatings with high quality. Then, the adhesion of top diamond coatings were estimated using Rockwell C indentation analysis, revealing that the adhesion of top diamond coatings on the WC-6%Co substrates can be improved by the interlayers with the diamond/ $\beta$ -SiC composite structures. Comprehensive TEM interfacial analysis exhibits that the cobalt diffusion is weak from WC-6%Co substrate to the composite interlayer. The homogeneous microcrystalline diamond coatings with the most excellent adhesion can be fabricated on the substrates with the composite interlayer with the  $\beta$ -SiC/diamond ratio of about 45%. The composite structures are appropriate for the application in high-efficiency mechanical tool as a buffer layer for the deposition of the diamond coating.

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### 1. Introduction

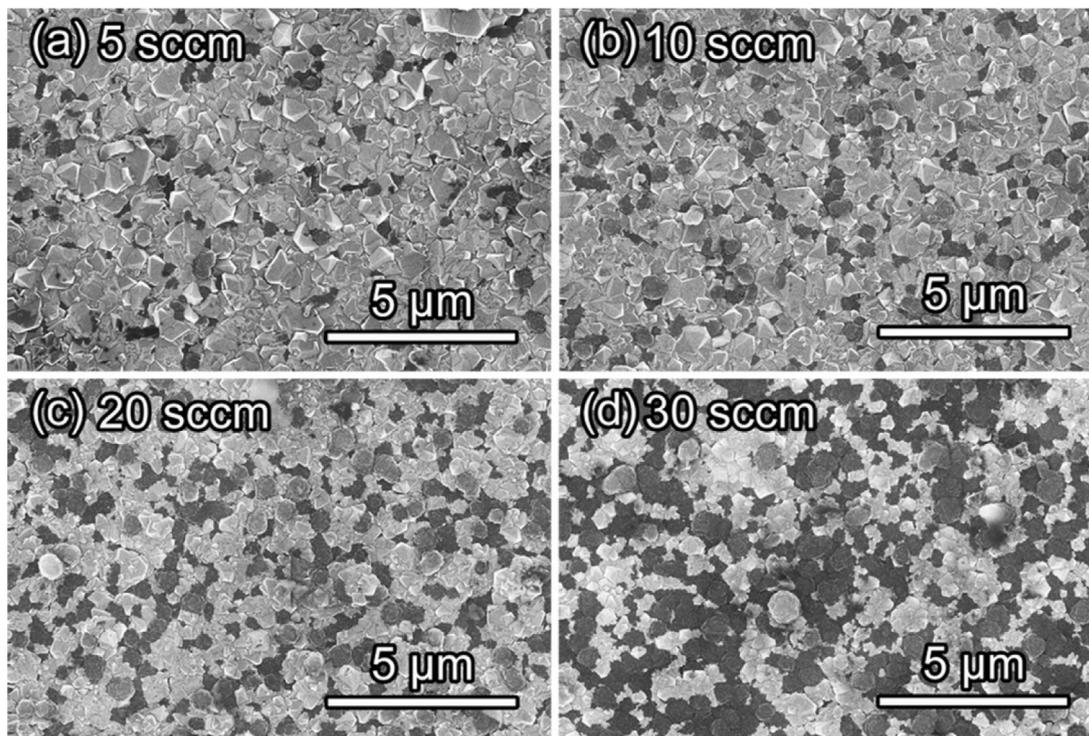
Diamond coating grown by chemical vapor deposition (CVD) processes onto cemented WC-6%Co substrates is an ideal candidate for machining applications owing to its extreme hardness, low friction coefficient, and chemical stability. CVD diamond coated tools have significant advantages considering the cost and fabrication flexibility compared to synthetic polycrystalline diamond (PCD) tools [1]. However, the performance of CVD diamond coated

tool is limited induced by the weak coating-substrate adhesion [2], owing to the strong catalytic effect of the cobalt from WC-6%Co substrate to the top diamond coating. Thus, the service life of diamond coating is determined by the coating delamination [3], rather than its own wear. To solve this problem, several approaches have been developed to prevent the cobalt diffusion into the diamond coating [4,5], categorizing into two types: the removal of cobalt binder using surface pretreatment and the introduction of an interlayer deposited between the top diamond coating and the substrate [6,7]. The acid etching, as the most important surface pretreatment, has been widely used to treat the cemented carbide tools [8]. However, surface toughness of the substrate is decreased after the etching process. Furthermore, the residual cobalt inside the substrate can gradually diffuse to the etching surface at the

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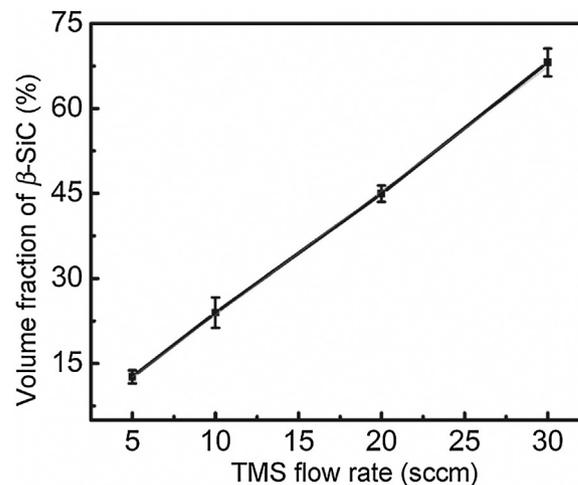
<sup>1</sup> These authors contributed equally to this work.



**Fig. 1.** FE-SEM micrographs of diamond/ $\beta$ -SiC composite interlayers deposited with different TMS flow rates: (a) 5 sccm; (b) 10 sccm; (c) 20 sccm; (d) 30 sccm. The flow rates of  $H_2$  and  $CH_4$  were kept at 400 sccm and 4 sccm, respectively.

high temperature during the diamond deposition [9]. The deposition time and thickness of diamond film are limited. Therefore, introduction of intermediate layers are normally used to resolve the problems [10,11]. Up to now, varies barriers, ranging from metals to ceramics, such as tungsten [12], aluminum [13], chrome [14], NbC [15],  $Si_3N_4$  [16], TiN, Ti(C,N) and CrN [15,17,18] by the physical vapor deposition (PVD) or CVD deposition to prevent cobalt diffusion are suitable choices [19]. However, these intermediate layers are not still satisfactory due to the low diamond nucleation density and growth rate during the subsequently top diamond deposition. The samples are normally to be pretreated in order to increase the diamond nucleation density [20], which added the complexity of the whole industry process.

Owing to the thermal expansion coefficient of  $\beta$ -SiC ( $3.8 \times 10^{-6} K^{-1}$ ) intermediating between diamond ( $1 \times 10^{-6} K^{-1}$ ) and WC-6%Co (Co 6 wt%,  $4.5 \times 10^{-6} K^{-1}$ ) substrate, diamond/ $\beta$ -SiC composite film as a gradient composite interlayer to alter the stress distribution was deposited to reduce the thermally induced shear stress [21]. The stress distribution is tunable by adjusting the  $\beta$ -SiC ratio in the composite film, which can be controlled by changing the component in the gas-phase reactions. Recently, it was reported that SiC was deposited for strengthening the adhesion between diamond particles and graphite substrate [22]. In our previous work [23–25], diamond/ $\beta$ -SiC composite interlayers were deposited on Si, W and Mo substrates by microwave plasma chemical vapor deposition. Furthermore, the stress-free diamond coatings were deposited on the Si substrate by employing the diamond/ $\beta$ -SiC composite interlayers [26]. In our previous reports, attention has been paid to the detailed structure of diamond and  $\beta$ -SiC in the composite film deposited on the Si substrate [27–29]. However, when the WC-6%Co is selected as the deposition substrate, the effect of cobalt diffusion from WC-6%Co substrate to top diamond coating is inevitable and the crystallinity of top diamond coating decreases [30,31]. The composite film enhanced the binding strength between diamond coating and substrate



**Fig. 2.** Plot of  $\beta$ -SiC volume fractions in composite interlayers as a function of TMS flow rates.

while the surface cobalt catalytic effect is weakened [30]. Here, on the basis of our previous work, we deep understanding about the growth of diamond/ $\beta$ -SiC composite interlayers on WC-6%Co substrates. The detailed microstructures and the diffusion of Co of the composite interlayers on WC-6%Co substrates as a function of TMS flow rates were investigated by FE-SEM, XRD, XPS, EPMA and TEM. The well adherent and effective composite interlayers between diamond and WC-6%Co substrate were used for diamond coatings, characterized by the Rockwell C indentation tests. The improvement of adhesion property for the composites interfaces on the diamond films was discussed in this work.

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