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Microstructure and properties of diamond/SiC composites prepared by tape-casting and chemical vapor infiltration process

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Abstract

This article reported a novel method for preparing diamond/SiC composites by tape-casting and chemical vapor infiltration (CVI) process, and the advantages of this method were discussed. The diamond particle was proved to be thermally stable under CVI conditions and the CVI diamond/SiC composites only contained diamond and CVI-SiC phases. The SEM and TEM results showed a strong interfacial bonding existed between diamond and CVI-SiC matrix. Due to the strong bonding, the surface HRA hardness could reach up to 98.4 (HV 50 ± 5 GPa) and the thermal conductivity (TC) of composites was five times higher than that of pure CVI-SiC matrix. Additionally, the effects of diamond particle size on microstructure and properties of composites were also investigated. With the increasing of particle size, the density and TC of composites with the size 27 μm reached 2.940 g/cm^3 and 82 W/(m K), respectively.

Keywords: CVI; Tape-casting; Diamond/SiC composites; Mechanical and thermal properties

1. Introduction

In recent years, due to its high thermal conductivity (TC), low coefficient of thermal expansion (CTE), ultrahigh hardness, high wear resistance and other outstanding properties, diamond/SiC composites are arousing great interests in many areas of industrial applications, especially the polycrystalline diamond compact (PDC) bits, electronic packaging materials, ultra stable substrate of optical mirror, and cutting tools.[1,4,6–9,11]

Up to now, the preparation of bulk diamond/SiC composites was mainly focused on sintering methods.[1–13] For sintering a mixture of diamond and SiC powder, high pressure and high temperature (HPHT) were needed to improve the density of composites and inhibit the graphitization of diamond.[1–3,14] However, considering the strict experiment conditions of HPHT, it was difficult to produce bulk or complex shaped composites. Besides, the graphitization of diamond during HPHT sintering could hardly be avoided without metal additives. There were also some other researchers choosing hot isostatic pressing (HIP) sintering, spark sintering (SPS) and silicon infiltration (SI) sintering. Generally, the raw materials for HIP, SPS and SI methods were diamond, amorphous carbon and silicon powders. And SiC matrix was commonly formed in situ by silicon diffusion–reaction mechanism.[12,13] Therefore, the residual silicon in composites was inevitable and that would affect the mechanical and thermal properties of composites especially at high temperature. In both Osamu and Masaru's researches,[5,6] graphite was not detected in the HIP products although the applied pressure was quite lower than that of the diamond stability field. In addition, more complex shaped and larger sized HIP products could be obtained. As to the SPS process with higher heating rate and shortened dwelling time, it is the fastest among all sintering methods until now.[8] Compared with HIP and SPS methods, SI was more commonly used. And its development was from high or normal pressure reactive molten infiltration (RMI) to low-pressure reactive vapor infiltration (RVI).[8–13]

Taking the above into account, diamond particle was unstable under some sintering conditions and its graphitization problem could not be fully solved. These, especially graphitization, would form weak phases in these composites and had a bad influence on the hardness and abrasion properties.[1,2,15] Besides,

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when the composites containing residual silicon were employed at a temperature approaching or over 1400 °C, they would suffer a destroying damage with the residual silicon turning soft and melting finally. Therefore, a new preparation method should be developed, which could not only maintain the thermal stability of diamond but also introduce no silicon into products. As we all know, chemical vapor infiltration (CVI) is a relatively mild process for its low deposition temperature and has been widely used to prepare ceramic matrix composites (CMCs).[16,17] Based on the Berman-Simon line theory,[18] diamond particles can still keep stable under the CVI temperature around 1000 °C and the graphitization of diamond can be avoided simultaneously. Meanwhile, compared with HIP, SPS, RVI, and RMI techniques, CVI-SiC matrix is produced by the in situ chemical vapor deposition (CVD) on reinforcements' surface. Consequently, the residual silicon in composites could be fully eliminated during CVI process. However, there usually existed a density gradient along the CVI bulk composites,[17,19,20] and generally, smaller the thickness of preforms was, higher the density of composites was. So taking advantage of tape-casting technique, porous preforms made of diamond particles with a thickness ranging from 300 to 500 μm could be prepared. And in order to test some mechanical and thermal properties of composites, we adopted a cyclic process of tape-casting and CVI on the substrate of CVI diamond/SiC composite until the thickness of samples met testing requirement of 3 mm.

In this paper, conventional methods were replaced by tape-casting and CVI to fabricate diamond/SiC composites, and the advantages of this method was also discussed. Microstructures of CVI diamond/SiC composites were observed, the phase composition and its stability were studied, and then the effects of diamond particle sizes on densities, mechanical and thermal properties of CVI diamond/SiC composites were also investigated.

2. Experimental procedure

2.1. Raw materials

The parameters (provided by manufacturer) of diamond powders (purity ≥99.6%, Hnboreas Ltd, China) are in Table 1, four grades of diamond powders with mean sizes of 5.2, 8.5, 16.5 and 27 μm were used in this experiment, which were marked as P5.2, P8.5, P16.5 and P27, respectively. Polyvinyl butyral (PVB, purity 99%, Haocheng Ltd, China) was used as binder and pore former, triethyl phosphate (purity 99.85%, Fuchen Ltd, China) were used as dispersant, glycerol (purity 99%, Fuchen Ltd, China) and dioctyl phthalate (DOP, purity 99.5%, Tianli Ltd, China) were used as plasticizer, isopropanol (purity 99.7%, Fuyu Ltd, China) and toluene (purity 99.7%, Hongyan Ltd, China) were used as solvent.

Table 1
The parameters of diamond powders used in the experiment.

Diamond grade	Mean particle size (μm)	Sphericity (%)
P5.2	5.2	60.17
P8.5	8.5	59.34
P16.5	16.5	73.28
P27	27	88.44

2.2. Preparation process

The preparation process sketch for laminated diamond/SiC composites is shown in Fig. 1. Firstly, the slurry of diamond particles was prepared through two steps by mixing in plastic containers. The first step was the dispersion of diamond particles with triethyl phosphate and isopropanol/toluene which were mixed for 7 h. The second step was adding binder (PVB), and plasticizer (glycerol/DOP) to the diamond particles dispersions for anther 7 h mixing. The relative weight radio of PVB to diamond was 1:15. The mixing speed was 120 rpm. Secondly, the slurry was degassed in vacuum to remove gas bubbles, and then a sheet made of diamond particles with a thickness around 300 μm was prepared by tape-casting. Thirdly, CVI diamond/SiC composites substrate was prepared from the sheet after CVI process. In CVI process, Methyltrichlorosilane (MTS: CH_3SiCl_3), hydrogen and argon were used as a precursor, carrier and dilute gas, respectively. The deposition conditions were as follows: $MTS/H_2 = 1/10$, $P = 5$ kPa, Ar = 350 ml/min, $t = 80$ h and $T = 1000$ °C. At last, diamond/SiC composites were fabricated by alternately tape-casting and CVI process when the thickness of composites reached up to 3 mm.

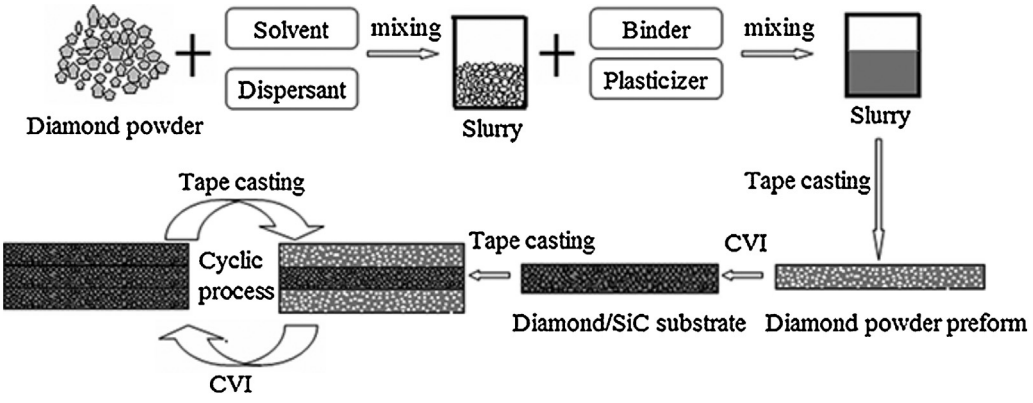

Fig. 1. Preparation process sketch for laminated CVI diamond/SiC composites.