ELSEVIER

Materials Science and Engineering A 428 (2006) 346–350

MATERIALS SCIENCE & ENGINEERING A

www.elsevier.com/locate/msea

Silicon carbide whisker reinforced silicon carbide composites by chemical vapor infiltration

Yunfeng Hua *, Litong Zhang, Laifei Cheng, Jing Wang

National Key Laboratory of Thermostructure Composite Materials, Northwestern Polytechnical University, Xi'an 710072, PR China

Received 21 February 2006; received in revised form 23 April 2006; accepted 16 May 2006

Abstract

Silicon carbide (SiC) whisker reinforced SiC ceramic matrix composites were prepared by chemical vapor infiltration. Microstructure and mechanical properties of the composites were investigated. The fracture toughness of the composites was improved to 5.65–7.21 MPa $m^{1/2}$, and its flexural strength value was between 196 MPa and 305 MPa, depending on the relative density. The pullout of rough whisker, crack deflection and whisker bridging are responsible for the improvement in fracture toughness. The randomly oriented whiskers that paralleled to the crack plane, and the weak bonding strength between whisker and matrix could not have much stress transfer from the matrix, leading to the low flexural strength.

Keywords: Chemical vapor infiltration; Organic binder; Reinforcement clusters; Preform; SiC_W/SiC

1. Introduction

Silicon carbide (SiC) is one of the promising candidate ceramic materials for a variety of high temperature, high stress and severe erosion applications in aerospace, hot engine and energy conversion devices because of its excellent high temperature properties. However, its low fracture toughness (K_{IC}) has long hindered this material from being used for wide applications. One major research direction has been to increase its fracture toughness by changing microstructures through the incorporation of other materials, and substantial toughening with isotropic property can be achieved by introducing whisker [1,2].

The conventional methods for preparation of whisker reinforced ceramic matrix composites are sintering, often involving aids. The whisker can be damaged mechanically during ball milling dispersion, and it also can be damaged chemically via a solution and reprecipitation process during the densification because of the liquid sintering aids. The toughening effect of the whiskers can be decreased due to its damage [2–5]. The residue of sintering aids exists as continuous intergranular glassy phase, which will soften and volatilize at elevated temperature. And microcracks also can be induced by thermal stress due to the mismatch of matrix, intergranular phase and heterogeneous reinforcements. Both of them have a negative effect on high temperature mechanical and chemical properties of composites [6–8]. Incorporation of other materials has been accompanied by the degradation of desirable properties of SiC. Although isotropic SiC whisker reinforced SiC matrix composites (SiC_W/SiC) with minor sintering aids can be prepared by hot isostatic pressing, it needs very high temperature and pressure and ultra-fine and high purity powders with the drawback of technological difficulties in manufacturing as well as with respect to costs, and the SiC whiskers with high volume fraction and large aspect ratio are difficult to be introduced into the SiC matrix. The reduction in whisker aspect ratio and the extensive growth of grains can mask any benefit the whisker has imparted [9–11]. The contribution of whisker on toughening also can be substantially decreased by the strong whisker–matrix bonding strength due to sintering, and appropriately strong bonding can facilitate the debonding at the whisker–matrix interface and activate the wake-toughening [12–14].

Based on these research results, we have investigated the possibility for improving the fracture toughness of the SiC_W/SiC composites by increasing whisker volume fraction and aspect ratio and improving working temperature by avoiding sintering aids. Chemical vapor infiltration (CVI) has been demonstrated to be an effective and mature enough method to prepare SiC matrix with ultra-pure and controllable grain sizes [15–20]. In this study, SiC_W/SiC is prepared by depositing SiC matrix on the internal surfaces of the porous SiC whisker preforms by CVI,

* Corresponding author. Tel.: +86 29 88486068x827; fax: +86 29 88494620.
E-mail address: nanojing@163.com (Y. Hua).

0921-5093/$ – see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.msea.2006.05.050

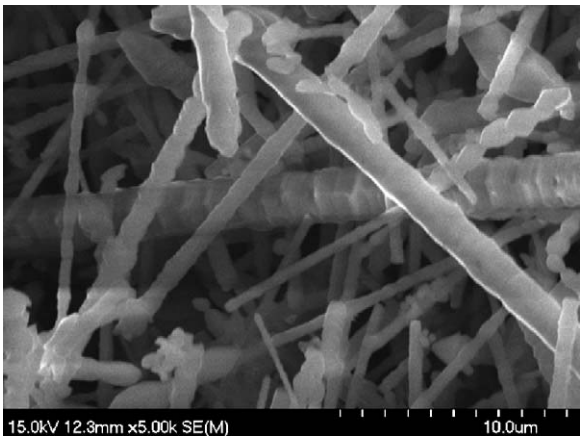

Fig. 1. SEM micrograph of the silicon carbide whiskers.

and the purpose of this work is to evaluate the employment of CVI, as a novel method, in fabricating the SiC_W/SiC from SiC whisker preforms, and its microstructure and mechanical properties.

2. Experimental procedure

Fig. 1 shows the SEM micrograph of the SiC whisker with rough surface. β-SiC whisker (99% pure, Alfa Aesar, MA, USA) had an average size of 18 μm in length and 1.5 μm in diameter. However, SiC whiskers as long as 50–100 μm were observed.

The procedure for whisker surface treatment was as follows. As-purchased SiC whiskers were first washed with concentrated hydrochloric acid (36%) to remove metallic impurities. After repeatedly washing with distilled water, the whisker was dipped into 20% hydrofluoric acid to remove SiO_2, and washed again with distilled water. Then, after drying, it was dipped into acetylacetonate to pre-disperse for 24 h. Then the solvent was extracted by a rotary evaporator. As-treated whisker was then stirred into the 10% acetone solution of epoxy resin and polyimide, and mixed using heavy duty laboratory mixer (Silverson Machine Ltd., Model L2R) for 24 h. After faster drying to maintain the uniformly dispersed state, the resultant SiC whisker was packed in a stainless steel die, and cold-pressed at room temperature and 30 MPa. The green body was then crushed and screened to select clusters with size of 0.3–0.6 mm. Again, the selected clusters were mixed with 5% water solution of poly-vinyl alcohol, and cold-pressed at room temperature and 15 MPa to mold the preforms. The volume fraction of the whiskers was about 25 vol.%. After burning out the organic, CVI was performed to deposit SiC matrix from CH_3SiCl_3 (MTS)/H_2 = 1/10 for 300 h at P = 3 kPa, Ar = 350 ml/min and T = 1100 °C. The prepared composites were machined, and cut into specimens with dimension 3 mm × 4 mm × 40 mm to further deposit SiC matrix for 50 h, 100 h and 150 h, respectively.

Specific mass was determined according to Archimedes method. The phase analysis of SiC whisker, SiC matrix and SiC_W/SiC composites was identified by X-ray diffraction (XRD) and microstructure was studied using scanning electron microscopy (SEM). Bend specimens with dimension 3 mm × 4 mm × 40 mm were used to evaluate the three-point flexural strength using a cross-head speed of 0.5 mm min^{-1} and a span length of 30 mm at room temperature. Five specimens were tested to obtain an average value. Micro-hardness and fracture toughness were determined at room temperature by Vickers indentation method with 196 N load for 30 s, 10 times on each of five samples per data point.

3. Results and discussion

3.1. Phase analysis and density

The relative density of 25 vol.% SiC_W/SiC composite to deposition time is summarized in Table 1. As shown, the density of the specimens is low compared to that of the whisker reinforced ceramic matrix composites prepared by sintering method [9,11]. CVI process results in composites intrinsically display significant residual porosity (typically, about 10%), which is the major factor inducing the low density [15]. Fig. 2 shows the XRD patterns of SiC whisker, pure SiC matrix and composites with deposition time of 450 h. As can be seen, the content of high temperature α-SiC phase in the composites increases, and the composites consist of pure SiC.

Dispersing and large aspect ratio of whiskers can create enough space in clusters. The spacing structure among and in the clusters is similar to that of the fiber preforms, creating path for the CH_3SiCl_3 and H_2 to infiltrate. After depositing SiC matrix for 450 h, the SiC coating (shell) still does not form, and the relative density of the composites is 88.72%. The relative density of 88.72% is close to its limit that can be achieved by CVI. It

Table 1
Deposition time and relative density of the 25% SiC_W/SiC composites

Deposition time (h)	Relative density (%)
300	84.30
350	87.95
400	88.40
450	88.72

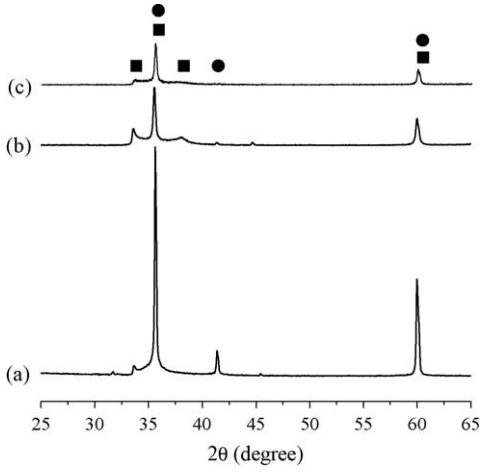

Fig. 2. XRD patterns of: (a) SiC whisker, (b) SiC_W/SiC composites and (c) CVI SiC. (■) α-SiC and (●) β-SiC.