JOURNAL OF MATERIALS SCIENCE **37** (2002) 2587–2590

Silicon nitride matrix composites with unidirectional silicon carbide whisker reinforcement

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SiC whisker reinforced Si_3N_4 was fabricated by fiber extrusion and hot pressing. SiC whiskers were unidirectionally oriented in a carrier fiber. The fibers containing the oriented whiskers were hot pressed in Si_3N_4 powder to form a SiC_w/Si_3N_4 composite with approximately 5 volume% whiskers. SEM micrographs were image processed to quantify whisker orientations in the extruded fiber and the composite. Oriented whiskers contributed to nominal increase in fracture strength over monolithic samples before and after thermal shock testing from 500, 600 and 700°C. *© 2002 Kluwer Academic Publishers*

1. Introduction

Silicon nitride (Si_3N_4) ceramics are receiving considerable research attention due to their high strength at high temperature, good thermal shock resistance and good oxidation resistance. However, these monolithic ceramic materials have some fundamental limitation in high temperature application because of brittle, catastrophic failure. Improvement in fracture toughness would expand the range of applications.

Whisker reinforcement had been identified as one of the major methods to toughen Si_3N_4 ceramics and improve their reliability [1–3]. Crack bridging and crack deflection are considered to be the main mechanism in whiskers toughening ceramics. Goto [4] pointed out one of the most important factors governing the effectiveness of these toughening mechanism was whisker orientation. Specimens with whisker orientations perpendicular to the direction of crack propagation will increase the fracture strength and creep resistance at high temperature. When the whisker orientation was parallel to the crack propagation direction, fracture strength decreased. Recently Wang [5] investigated the relationship between toughening action and whisker orientation in whisker reinforced ceramic composites. Whiskers would have a maximum toughening effect only in the case of a small orientation angle and weak interface where whisker pull- out would occur.

The objective of this paper was to develop and improve the properties of silicon nitride composite in a given direction by eliminating all the whiskers in the direction that would be detrimental to design properties. Extrusion and hot pressing were used as the processing methods. The evaluation of whisker orientation was done by Scanning Electron Microscopy (SEM).

2. Experimental procedure

The matrix powders containing a commercial grade Si_3N_4 (Ube Industries, Ltd.) and 3% MgO sintering additive were prepared by ball milling in methanol for 3 hours. A dry blend of SiC whiskers (Silar grade SC-9), cellulose derivative and Si_3N_4 matrix powder were mixed with acetone based on the following composition (Table I):

The slurry was extruded with a compressed air extruder with filament diameter approximately 0.26 mm. After partial burnout the organic material, the filaments were placed between Si_3N_4 matrix powders in unidirectional array in the graphite mold. This composite was hot pressed at 1670°C with 4000 psi for 1 hour and 15 minutes under vacuum. The SiC whisker content in the composite was under 5%. A monolithic Si_3N_4 sample was also hot pressed under the same process condition as a control.

The density of specimens was determined by the immersion technique. The microstructure and whisker orientation was observed with SEM. SEM image analysis was also performed in order to evaluate the degree of orientation. Bending strength was determined by three-point bend test and a water-quenching test was conducted in order to determine relative thermal shock resistance of the materials.

3. Results and discussion

3.1. Microstructure

Fig. 1 shows an SEM micrograph, which indicates a high degree of whisker orientation in the extruded cellulose base fiber. The oriented whiskers were oriented in a manner parallel to the extrusion direction of the fiber.

In order to quantify the whisker orientation, the positions of all visible fibers in Fig. 1 were traced by image processing. (Fig. 2)

The angle and length was measured of each traced whisker. The degree of orientation was measured in two ways. The first way involved counting the percent of whiskers oriented within five degrees. This showed

TABLE I Slurry composition of fiber extrusion

Material	wt%
Acetone	58.3
Cellulose derivative	21.7
SiC whiskers	13.4
Si_3N_4 matrix powder	6.6

Figure 1 SEM micrograph of whisker orientation in extruded fiber.

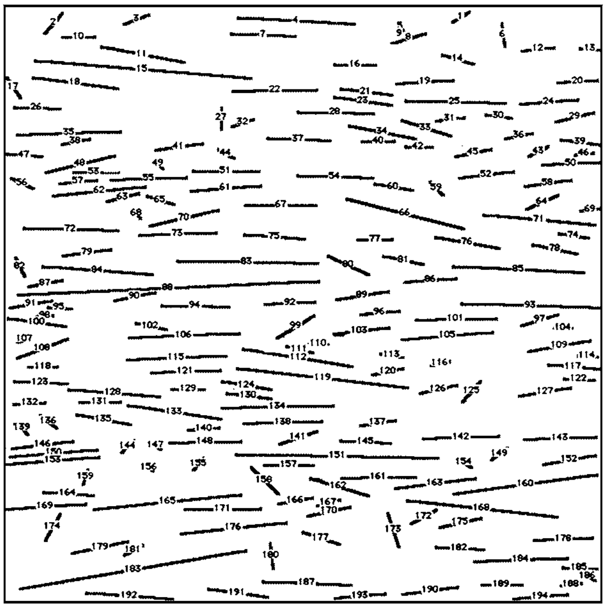

Figure 2 Image processed SEM micrograph of whisker orientation in extruded fiber (×2000).

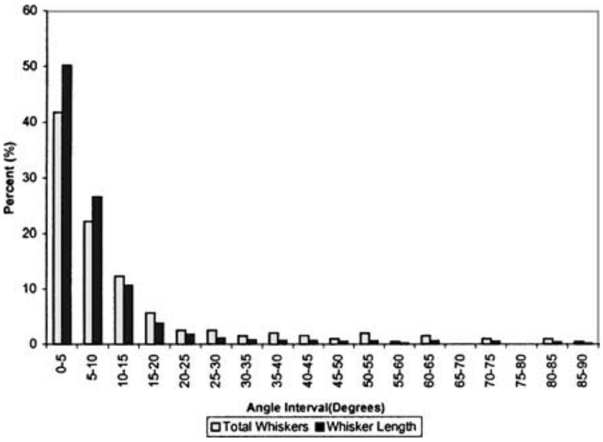

Figure 3 Percent whisker orientation of extruded fiber.

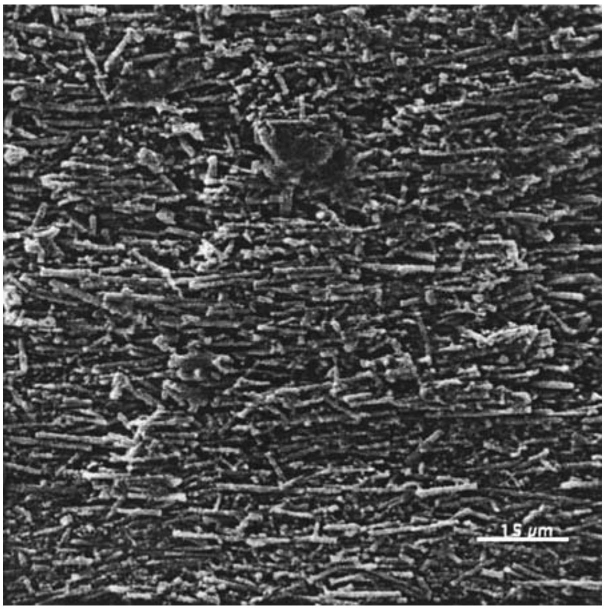

Figure 4 SEM micrograph of whisker orientation in SiC_w/Si_3N_4 composite.

the number of oriented whiskers. The second way was to sum the length of total whiskers and find the percent of total whisker length found within each interval (Fig. 3). While it is important to know the number of whiskers oriented along a particular angle, it is important to know the length of whiskers also. One misoriented long whisker can offset many oriented short whiskers.

In the work of Jia *et al.* [6], most of the whiskers in random distribution appeared to have an orientation of 45°. Half of the whiskers would be oriented between 0–45°, while the others would be oriented between 45°–90°. In this work, 63.9% of total whisker and 76% of total whisker length are found between 0–10° (Fig. 3), indicating that most of whiskers were oriented in a small angle distribution.

Fig. 4 shows the whisker orientation of the SiC_w/Si_3N_4 composite. The overall whisker orientation in the composite appeared better oriented than in the extruded fiber. The percent orientation was shown in Fig. 5. No whiskers were found beyond 45°. Between 0–10°, 81% of total whiskers and 85% of total whisker length were oriented. This view shows a high concentration of whiskers but the overall concentration was about 5% because of matrix phase separating the carrier filaments. The overall density of this composite is 3.06 g/cm^3, 96.3% theoretical density.

3.2. Mechanical properties

The flexural strength of Si_3N_4 monolithic and Si_3N_4-SiC whisker composites was tested with a three-point bend test.

Three test bars of each sample were broken in the three-point bend test. The results were given in Table II

TABLE II Room temperature flexural strength results

Material	(%) Theoretical density	SiC_w (%) Volume	Hot-pressed temperature °C	Flexural stress, σ MPa
Si_3N_4 Monolithic	97.46	0	1670	520
SiC_w/Si_3N_4 Composite	96.25	5	1670	680
Random SiC_w/Si_3N_4[6]	88	20	1800	470
Random SiC_w/Si_3N_4[6]	86	30	1800	450

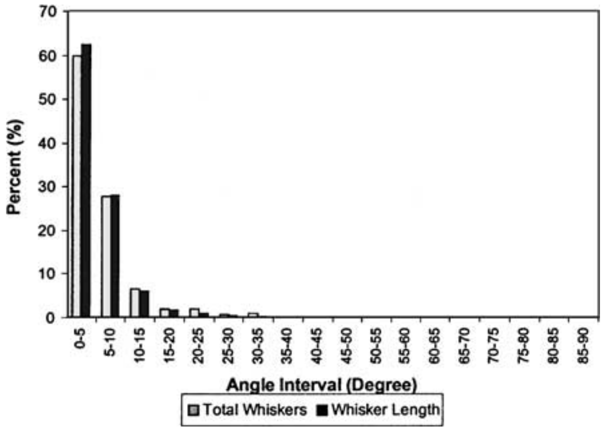

Figure 5 Percent whisker orientation of SiC_w/Si_3N_4 composite.

along with data provided from Reference 6 for random SiC whisker composites.

The oriented SiC_w/Si_3N_4 composite showed a somewhat higher value of flexural strength than the authors' monolithic material, which was also somewhat denser. This may be the result of strengthening by the oriented whiskers. Jia [6] reported that the bending strength of the randomly oriented SiC whiskers in Si_3N_4 composite decreases drastically with increasing SiC_w content. Data for some of his samples is also included in Table II. His explanation was that decreasing overall composite density offset any enhancement due to the increased whisker content. Another reason could be the random whisker orientation of these composites. Since SiC_w/Si_3N_4 composites usually display intergranular fracture, the bending strength of the composites is influenced by grain boundary phase as well as whisker orientation. In unidirectional whisker orientation, the bonding strength of grain boundary or interfacial phases may be a main factor influencing the strength of the composites. But in the random whisker orientation, whiskers that are parallel to the crack plane cannot have much stress transfer from the matrix, therefore the strength of composites drops significantly [4]. Whisker pull out was also observed in the fracture surface of the composite and it was believed that whisker pull out was the main mechanism of the strength improvement of these whisker-reinforced composites (Fig. 6).

A few bars of each sample were heated at 500°C, 600°C and 700°C for 15 minutes, then quenched in room temperature water.

TABLE III Thermal shock resistance parameter

Material	(%) Theoretical density	SiC_w Volume %	Thermal shock parameter (R) °C	Flexural stress, σ MPa
Si_3N_4 Monolithic	97.46	0	500	276
Si_3N_4 Monolithic	97.46	0	600	251
Si_3N_4 Monolithic	97.46	0	700	163
SiC_w/Si_3N_4 Composite	96.25	5	500	438
SiC_w/Si_3N_4 Composite	96.25	5	600	370
SiC_w/Si_3N_4 Composite	96.25	5	700	296
Random SiC_w/Si_3N_4[6]	88	20	500	452
Random SiC_w/Si_3N_4[6]	86	30	500	480

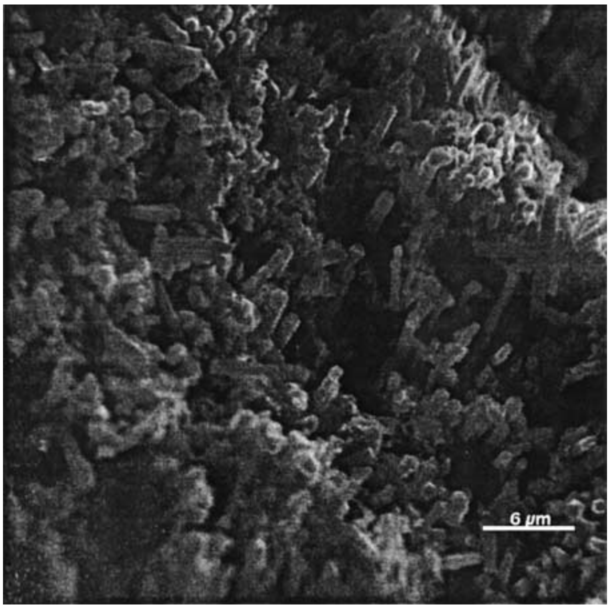

Figure 6 SEM micrograph of whisker pull out in the fracture surface of SiC_w/Si_3N_4 composite (×5000).

The monolithic sample showed a greater decrease in strength with increasing temperature gradient. The whisker-reinforced composites did not lose as much strength as the monolithic Si_3N_4 after this thermal shock test, because the whiskers in the composite material acted as barriers to extensive crack propagation, initiated by the thermal stress. In comparison, the data of Jia *et al.* [6] showed that the flexural strength of randomly oriented SiC_w composite after thermal shock testing showed higher values than the oriented SiC_w/Si_3N_4 composite of this investigation. This can be explained as follows. First, the content of SiC whiskers in SiC_w/Si_3N_4 composite (5-vol%) was less than in the random oriented SiC_w/Si_3N_4 (20 vol% or 30 vol%). As shown in Table III, the residual strength after thermal shock increased with increasing SiC_w concentrations due to the fact that more whiskers had the ability to deflect thermal stress cracks and also lower density was less shock sensitive. The whiskers keep the thermal shocked specimen under compression thus reducing the size of the surface flaws. So it is likely that the flexural strength of oriented whisker reinforced material after thermal shock will be comparable or even much better than the random one if the volume concentration of the oriented whiskers and the random oriented whisker are comparable in the composite material.

4. Conclusion

A process for providing highly oriented silicon carbide whiskers in a carrier filament of cellulose base material combined with additional matrix phase Si_3N_4 was successful.

Unidirectionally reinforced silicon nitride composite billets were prepared by hot pressing with additional Si_3N_4 matrix, which reduced the overall whisker concentration to 5 volume percent. Three-point bend strength testing showed values somewhat higher than comparable monolithic samples. Bending testing after exposure to sharp temperature gradients, induced by quenching from temperature ranging from 500 to 700°C, revealed a nominal decrease in residual strength for the monolithic samples as compared to the oriented whisker samples. Randomly oriented whisker composite samples with additional porosity with volume concentration of up to 30%, showed residual strengths comparable to the 5 vol% samples when exposed to 500°C shock parameter.

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Received 20 January 2000
and accepted 29 November 2001