Microstructure of Si_3N_4 Composites Reinforced with SiC Whiskers

Shigeki KOBAYASHI, Toshio KANDORI and Shigetaka WADA

(Toyota Central Research and Development Laboratories, Inc.)
(Nagakute, Aichi-gun 480-11)

SiC ウィスカーで強化した Si_3N_4 複合材の微細構造

小林茂樹・神取利男・和田重孝

((株)豊田中央研究所)

Si_3N_4 composites reinforced with SiC whiskers were fabricated using injection molding and HIP treatment. The strength and fracture toughness were significantly improved over monolithic Si_3N_4 . Microstructure of composites before and after HIP treatment was examined by transmission electron microscopy. No reaction layer was observed between whiskers and matrices. HIP treatment eliminated voids at the interface. The interfacial bonding was excellent and no pull-out of whiskers occurred on fracture. Factors controlling the improvement of properties are discussed based on the microstructural analysis.

[Received February 17, 1986]

Key-words : Silicon nitride, Silicon carbide whisker, Composite, Microstructure, TEM, Interface

Ceramics such as Si_3N_4 and SiC are promising materials for high temperature structural applications. Using ceramics in heat engine systems may bring the benefits of efficiency and fuel economy. The major obstacle for assuring the reliability of ceramics as structural components is the brittleness of ceramics or low fracture toughness. Ceramics matrix composites will give a solution for this problem. Although several pioneering studies have been made¹⁾, much effort is required to improve the properties of composites and to understand the behavior of composites.

In this note, the microstructural analysis of Si_3N_4 composites reinforced with SiC whiskers will be reported. Several new processes have been employed to fabricate the composites; i. e. the injection molding for aligning whiskers and HIP treatment for achieving dense matrices. The composites fabricated had superior properties over monolithic Si_3N_4 . The factors controlling the improvement of properties will be discussed based on the microstructural analysis.

The details of experimental procedures will be reported elsewhere²⁾. Powder mixture was prepared from Si_3N_4 , Y_2O_3 , MgAl_2O_4 powders and SiC whiskers (10 wt%). Test specimens made by injection molding were gas-pressure sintered in nitrogen of 1 MPa at 1700°C, followed by HIP

ing at 1500°-1900°C in nitrogen. Light microscope observation revealed that most of SiC whiskers are aligned along the flow direction during injection molding, which is the longitudinal direction of test specimens. An addition of SiC whiskers retarded sintering process, resulting in lower bulk density (3.20 g/cm³). However, post-HIP'ing increased this to 3.21-3.23 g/cm³, depending on the HIP'ing temperature. With the selection of optimum HIP'ing temperature, the four-point flexural strength and fracture toughness measured by the chevron notch method could be increased up to 950 MPa and $9.8 \text{ MPa}\sqrt{m}$, respectively. These properties of the composites have been significantly improved compared with those of the monolithic Si_3N_4 and also the composites reported in literatures^{3),4)}.

Thin foils for transmission electron microscopy (TEM) observation were prepared from the cross section of the test specimens. After mechanical polishing, foils were ion-milled to perforation. A typical structure for HIP'ed composites is shown in Fig. 1. SiC whisker (C) is surrounded by many Si_3N_4 grains (N). The shape of whiskers is nearly hexagonal in axial view, as shown in Fig. 1 and Fig. 2. Analyses of diffraction pattern showed that the growing direction of whiskers is [0001] (hexagonal coordinates) or [111] (cubic

Fig.1. Microstructure of composites after HIP treatment. SiC whiskers (C) are surrounded by many Si₃N₄ grains (N). The shape of whiskers is nearly hexagonal and the growing direction is [0001] (hexagonal coordinates) or [111] (cubic coordinates).

coordinates) in most cases, which is consistent with other reports⁽⁵⁾⁻⁷⁾. As shown in Fig.2, whiskers have growing defects such as stacking faults and partial dislocations. Diffuse contrast in the core region of whiskers will be due to cavities as reported by Nutt⁽⁷⁾. Viewing inclined to whisker axis, the disorder array of stacking faults can be seen as shown in Fig.3. This is due to that there are hundreds of polytypes in SiC crystals and they are variants of a one-dimensional stacking sequence of closed-packed planes. These observations confirmed the thermal stability of SiC whiskers in Si₃N₄ matrix during sintering. This is certainly an important factor for the improvement of properties.

A striking difference can be noticed between the microstructure of composites before and after HIP'ing. Before HIP'ing, voids exist along the part of interface because sintering is retarded by SiC whisker addition (Fig.2). These voids are eliminated by HIP treatment and the entire periphery of whiskers contacts with Si₃N₄ matrix (Fig.1). This contributes to achieving higher density after HIP'ing. As shown in

Fig.2. Microstructure of composites before HIP treatment. Since an addition of SiC whiskers retarded sintering, voids (V) exist at the interface between whiskers (C) and Si₃N₄ matrix (N). Growing defects such as stacking faults and partial dislocations can be seen within the whisker.

Fig.3. Enlarged TEM picture showing the excellent bonding of the interface in the HIP'ed composites. No reaction layer exists between whiskers (C) and Si₃N₄ grains (N).

Fig. 3, SiC whiskers and Si_3N_4 grains contact each other without any reaction layer. The bonding between whiskers and Si_3N_4 grains is as good as that among Si_3N_4 grains, which may contribute to the improvement of strength.

In terms of fracture toughness, the interfacial bonding should be weak enough to allow the matrix cracks to propagate along the interface^{8),9)}, resulting in matrix-whisker debonding and whisker pull-out on fracture. Since the composites in this study have a strong interfacial bonding, the matrix cracks will propagate through whiskers, leading to brittle fracture. Fractographic observations showed that no pull-out of whiskers was detected and there was no significant difference on fracture surfaces between the composites and monolithic Si_3N_4 . Thus, at least for this composite system, the toughening mechanism of fiber debonding does not play an important role. Several other factors can be considered for a significant increase of fracture toughness.

The fracture toughness is related to the two material parameters; Young's modulus and fracture energy. The Young's modulus of composites is increased by adding SiC whiskers of

higher modulus. Higher fracture energy is expected by (1) the residual compressive stress due to the difference of thermal expansion coefficient between matrices and whiskers, and (2) strengthening of Si_3N_4 grains and grain boundary due to HIP treatment. The strength of composites is controlled by two factors; i. e. fracture toughness and flaw size. The improvement of strength after HIP'ing will be largely due to an increase in fracture toughness. Further study is needed to pinpoint the controlling factors of toughening and also to optimize the properties of composites by tailoring the interfacial bond.

References

- 1) See *Ceram. Eng. and Sci. Proc.*, 5, No.7/8 (1984).
- 2) T. Kandori, T. Honma and S. Wada, Proc. Annual Meet. Ceram. Soc. Japan (1986).
- 3) K. Ueno, *Bull. Ceram. Soc. Japan*, 18, 1040-46 (1983).
- 4) G. C. Wei and P. F. Becher, *Am. Ceram. Soc. Bull.*, 64, 298-304 (1985).
- 5) L. I. van Torne, *J. Appl. Phys.*, 37, 1849-51 (1966).
- 6) J. J. Comer, *Mater. Res. Bull.*, 4, 279-87 (1969).
- 7) S. R. Nutt, *J. Am. Ceram. Soc.*, 67, 428-31 (1984).
- 8) E. Fitzer, Proc. Int. Symp. Factors in Densification and Sintering of Oxide and Non-Oxide Ceramics Japan (1978) p.618-73.
- 9) B. A. Bender et al., *Ceram. Eng. and Sci. Proc.*, 5, 513-29 (1984).