Effect of whisker surface treatment on the mechanical properties of 20vol%SiCw/BAS glass-ceramic composites

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20vol%SiCw/BAS (barium aluminosilicate) composites with as-received, acid-leached and heat-treated whiskers were fabricated by a hot-pressing technique and their mechanical properties were evaluated. The whiskers used in these composites were characterized using Auger electron spectrometry (AES) and scanning electron microscopy (SEM), and the observations were correlated with the mechanical properties. The results show that the oxygen content of the whisker surfaces was reduced by the hydrogen fluoride (HF) acid-leaching treatment and the surface geometry of whiskers was smoothed by the heat-treatment in an inert gas atmosphere. The corresponding composites show an obvious increase (18% and 31%, respectively) in toughness compared with the composites reinforced with as-received whiskers. Fracture surfaces revealed evidence of toughening by the mechanisms of crack deflection, crack bridging and whisker pullout for the composites with surface treated whiskers. © *2001 Kluwer Academic Publishers*

1. Introduction

Ceramic matrix composites reinforced with whiskers are prospective materials for high-temperature structural applications. Glass-ceramic matrices are of particular interest because of their high melting temperature, low thermal expansion, oxidation resistance and low dielectric constant.

Barium aluminosilicate ($BaO{\cdot}Al_2O_3{\cdot}2SiO_2$) has one of the highest melting temperatures, 1760°C, among the glass-ceramic materials [1] and the monoclinic form exhibits a low thermal expansion coefficient (2.3×10^{-6}/°C from 22 to 1000°C) [2]. However, like other glass-ceramic materials, Barium aluminosilicate (BAS) glass-ceramic matrix exhibits relatively lower mechanical properties and hence limit its use in many structural applications [3, 4]. To improve the mechanical properties, it is desirable that some reinforcing agents are incorporated into the BAS matrix.

It has been reported that the strength and toughness of glass-ceramics can be greatly increased by the addition of SiC whiskers [5–9]. The increased fracture toughness of these composites is the result of crack interaction with the whiskers in the matrix and the main mechanisms are crack deflection, crack bridging and whisker pullout. The operation of these toughening mechanisms to a large extent depend on the nature of the whisker/matrix interface, which includes matrix chemistry, whisker surface chemistry and whisker morphology [5, 10–13]

In this paper, SiC whiskers were leached with HF acid and heat-treated in Ar inert gas atmosphere before being incorporated into BAS matrix to investigate the effects of surface treatment of SiC whiskers on the mechanical properties of BAS composites.

2. Experimental

The matrix used in this study was BAS glass-ceramic prepared by sol-gel process using alkoxide of Ba, Al and Si [4]. The β-SiC whiskers were supplied by the Shanghai Institute of Ceramics, having a diameter of 0.5–1 μm and a length of 30–100 μm. For the acid-leaching treatment, the whiskers were leached with 5vol%HF acid for 10 h and then washed five times with distilled water. The SiC whiskers were heat-treated at 1900°C in Ar gas atmosphere for 30 min in a graphic crucible.

The composites with both as-received and surface treated whiskers were hot-pressed at 1370°C for 30 min under a pressure of 10 MPa. The density of the samples was measured by Archimedes' method in distilled water at 20°C.

The flexural strength and the fracture toughness of the composites were measured in air at room temperature using an Instron-1196 machine. The flexural strength measurements were performed on bar specimens (3 mm × 4 mm × 36 mm) using a three-point bend fixture with a span of 30 mm. The fracture strength

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were performed on single edge-notched bar (SENB) specimens (2 mm × 4 mm × 25 mm) with a span of 16 mm and a half-thickness notch was made using a diamond wafering blade.

The surface chemistry of SiC whiskers was characterized by the AES technique. The surface morphology of SiC whiskers and the fracture surfaces of the composites were examined using a HITACHI-570 scanning electron microscope. An indentation load of 10 kg was used to introduce cracks within the specimens using a Vickers hardness tester and subsequently observing the crack propagation modes could be observed.

3. Results and discussion

Surface morphology of as-received and surface treated SiC whiskers are shown in Fig. 1, indicating that the surface of the as-received whiskers is very rough (Fig. 1a), and the surface heat treatment at high temperature make these surfaces rather smooth (Fig. 1c). It is accepted that a convex surface is greatly higher in chemical potential than a concave surface and hence mass tends to transfer from a convex to a concave surface during the heat-treating. The surfaces of the whiskers are smoothened by the heat treatment as a result of the above mass transfer effects. It was also found that the whisker size itself was not changed much after heat treatment and the β crystalline phase of whiskers remained unchanged after the heat treatment examined by XRD analysis. Akatsu [13] also found the similar effect of a surface heat treatment on the roughness of whiskers. The acid-leaching treatment also improves the surface morphology of the whiskers, but this effect is not obvious (Fig. 1c and d).

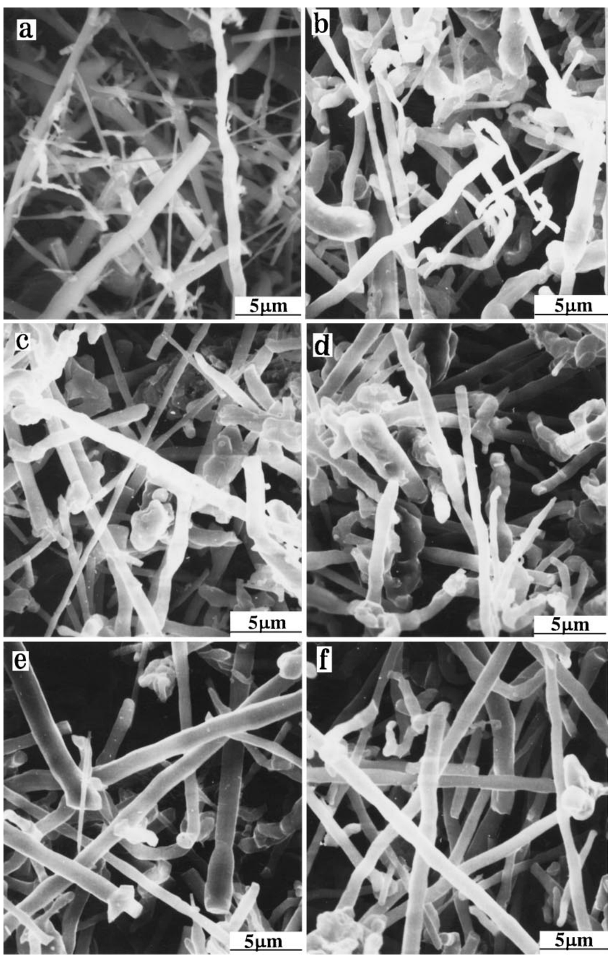

Figure 1 Morphologies of SiC whiskers (a), (b) as-received, (c), (d) acid-leached, (e), (f) heat-treated.

TABLE I Room temperature mechanical properties of 20vol%SiCw/BAS composites

Materials	Flexural strength, MPa	Fracture toughness, MPa m$^{1/2}$
SiCw(as-received)/BAS	272 ± 10	3.07 ± 0.25
SiCw(acid-leached)/BAS	270 ± 11	3.62 ± 0.23
SiCw(heat-treated)/BAS	253 ± 8	4.76 ± 0.26

The surface chemistry of SiC whiskers was obtained by Auger electron spectrometry and the results are shown in Fig. 2. It can be clearly seen that the amount of oxygen at the as-received SiC whisker surface is very high (28.34at%O). On the contrary, after acid-leaching treatment, these whiskers showed reduced surface oxygen content (2.3at%O), indicating that the acid-leaching treatment greatly removed the oxide on the whisker surfaces. The effect of the surface heat treatment on the chemistry of whisker surfaces is not obvious (Fig. 2c).

The improvement of changes in the surface chemistry and morphology of SiC whiskers undoubtedly has a strong influence on the nature of whisker/matrix interface and thus resulting the mechanical properties of the composites.

The mechanical properties of the composites are summarized in Fig. 3 and Table I. It can be seen that there is little change in flexural strength for the composites with different SiC whiskers. But the composites with surface treated whiskers showed an obvious increase in fracture toughness compared with the composites with as-received whiskers. The composites produced with acid-leached and heat-treated whiskers resulted in a fracture toughness increase of 0.55 and 1.14 MPa m$^{1/2}$, respectively.

The fracture surfaces of the composites tested in flexural strength testing are illustrated in Fig. 4, indicating that the fracture behaviors of the composites reinforced with different SiC whiskers are quite different. The composites with as-received SiC whiskers exhibited a relatively smooth fracture surface (Fig. 4a), with little evidenced of whisker pullout. However, incorporation of the surface treated SiC whiskers into the BAS matrix produced substantial crack/whisker interactions, with evidence of whisker pullout (Fig. 4b and c), especially for the composites with the heat treated whiskers.

The relationship between the crack path and the microstructure of the composites can be demonstrated more clearly by examining the crack propagating patterns produced from Vickers indentations, as shown in Fig. 5. For the composites with as-received whiskers (Fig. 5a), the crack always tends to propagate directly through the whiskers, exhibiting very little crack interaction with the microstructure and hence resulted a more planar fracture path. In contrast, for the composites with acid-leaching or heat treated whiskers, cracks

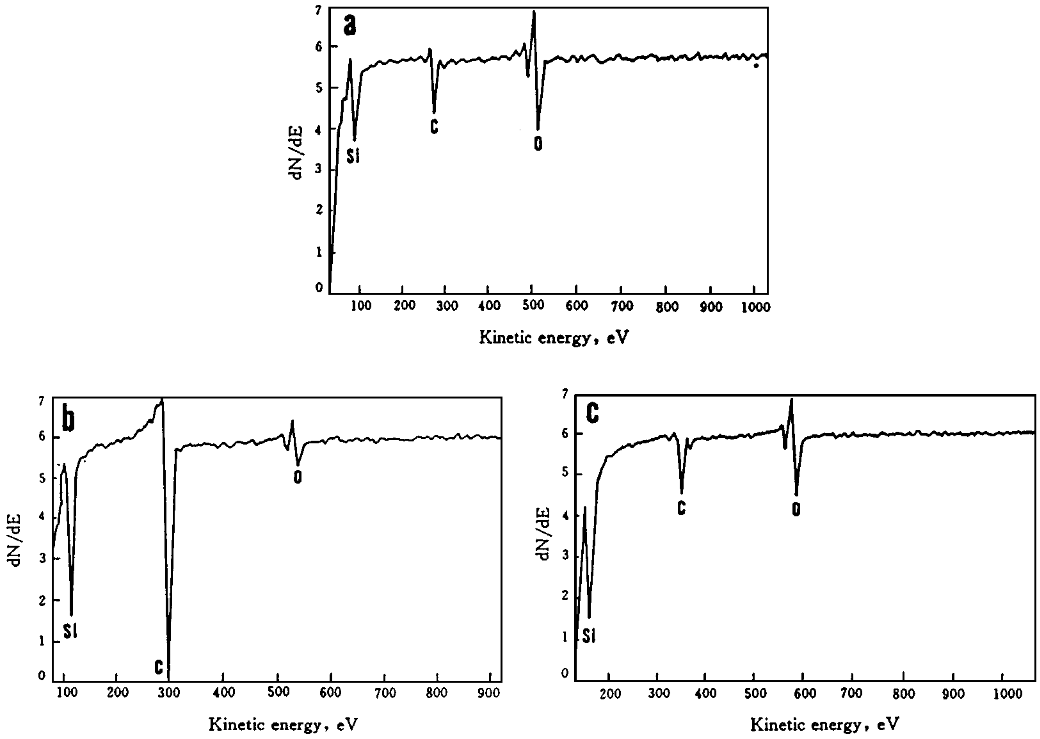

Figure 2 AES analysis of the surfaces of SiC whiskers. (a) as-received, (b) acid-leached, (c) heat-treated.

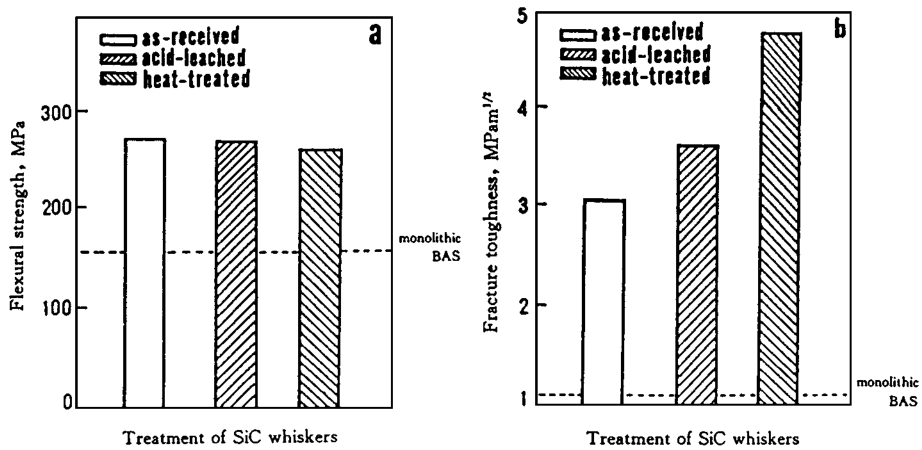

Figure 3 Effect of surface treatment of SiC whiskers on the mechanical properties of 20vol%/BAS composites (a) flexural strength, (b) fracture toughness.

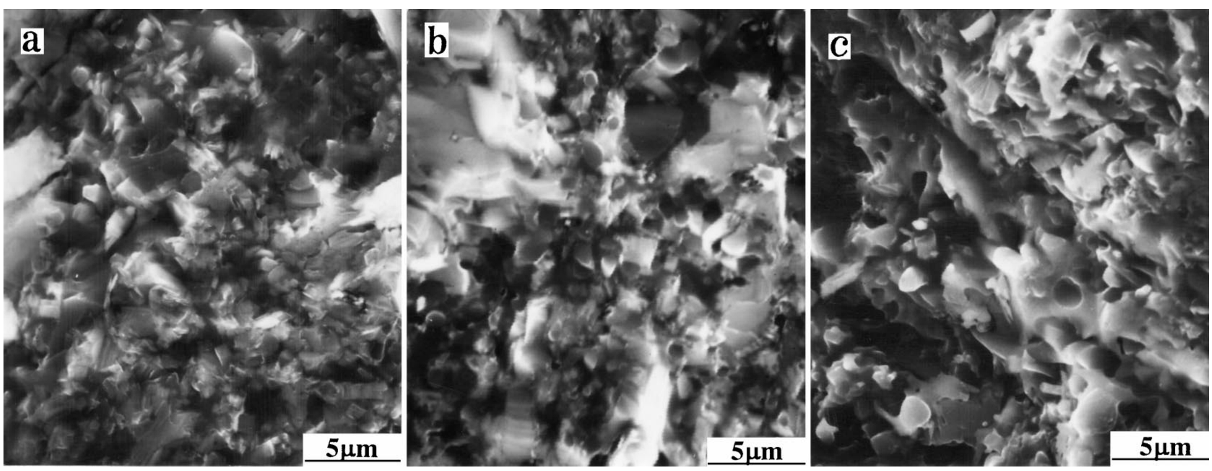

Figure 4 SEM photographs of bending fractured surfaces of 20vol%SiCw/BAS composites. (a) BAS composites with as-received whiskers, (b) BAS composites with acid-leached whiskers, (c) BAS composites with heat-treated whiskers.

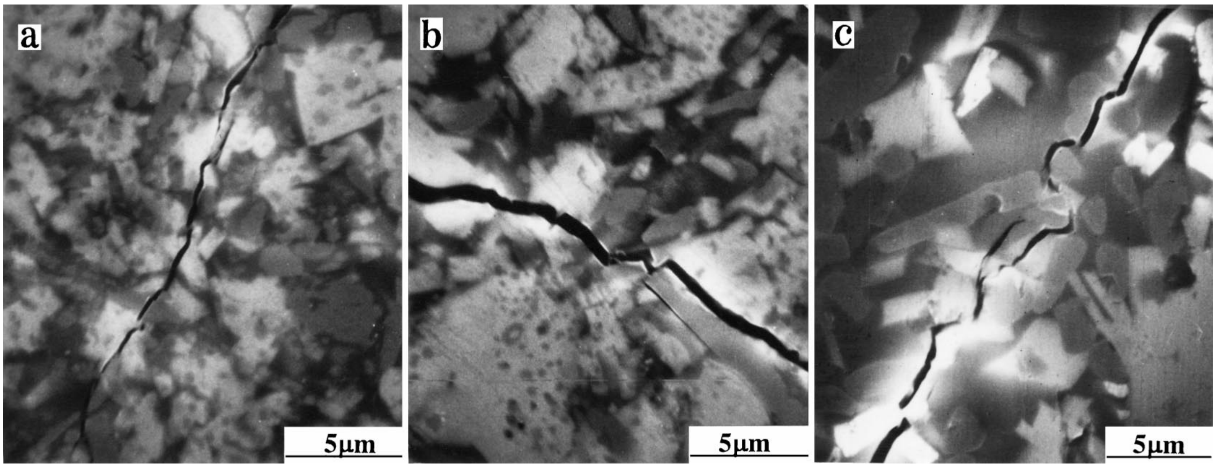

Figure 5 SEM photographs of 20vol%SiCw/BAS composites showing the propagation paths of cracks produced the corners of indentation during Vickers hardness. (a) BAS composite with as-received whisker, (b) BAS composite with acid-leached whiskers, (c) BAS composite with heat-treated whiskers.

were observed to deflect along the whisker/matrix interfaces (Fig. 5b and c), and the whiskers were observed to pullout and bridge in the wake of the extending cracks, resulting in a tortuous fracture surface. In addition, the composites with the heat-treated whiskers revealed a further increase in the extent of the crack/microstructure interactions.

The increase in fracture toughness of BAS composites reinforced surface treated whiskers is contributed to the improvement of the whisker/matrix interfacial structure. As shown in Fig. 1a, the as-received whiskers have rough surface, so that the frictional resistance at the interface is very large and causes whiskers fracture before pulling-out. On the other hand, the high surface SiO_2 contents of the as-received whiskers increased the strength of whisker/matrix bonding, which also inhibited the whiskers to be pulled out. The lack of whiskers pullout and planar fracture surfaces (Figs 4a and 5a) confirmed this.

The acid-leaching treatment minimized the whisker surfaces oxygen content (Fig. 2b) and hence resulted in a decrease in bonding strength at the whisker/matrix interface. This can be confirmed by the evidence of whisker pullout and crack deflection along the whisker/matrix interface (Figs 4b and 5b). Yang *et al.* [12] also reported that the acid-leaching treatment eliminated the SiO_2-rich layer presented on the surface of as-received whiskers and hence reduced the amorphous phase at the interface between the whisker and matrix, resulting in a reduction in fracture energy.

Treating the as-received SiC whiskers in Ar inert atmosphere smoothed the whisker surfaces by reducing the irregularities on them (Fig. 1e and f) and did not increase the whisker surface SiO_2 contents (Fig. 2c). This reduced the frictional resistance at the interface during whisker pull out and thereby facilitated it.

As stated above, reducing the whisker surface SiO_2 content and smoothening the whisker surface are very effective actions to further increase the toughening effect of whiskers.

4. Summary

The acid-leaching treatment can greatly reduce the whisker surface SiO_2 content and decreased the bonding strength of the interface between the SiC whiskers and BAS matrix. The heat treatment smoothened the whisker surfaces and resulted in a reduction of the friction resistance during whisker pulling-out. Either the decrease in the bonding strength of the whisker/matrix interface or friction resistance can promote whisker pullout during fracturing and hence further increase the fracture toughness of BAS glass-ceramic matrix composite.

References

1. C. H. DRUMMOND and W. E. LEE, *Ceram. Eng. Sci. Proc.* **10** (1989) 1485.
2. E. J. WUCHINA and I. G. TALMY, in Proceedings of the 14th conference on Metal, Carbon and Ceramic Composites, NASA Conf. Publ., 1990, Vol. 3079, Part I, p. 239.
3. C. H. DRUMMOND III, *Ceram. Eng. Proc.* **11** (1990) 1072.
4. F. YE, Post-doctoral research report, Northwestern Polytechnical University, People's Republic of China, 1997. In Chinese.
5. J. J. BRENNAN and S. R. NATT, *J. Am. Ceram. Soc.* **75** (1992) 1205.
6. H. W. JANG, K. S. KIM and C. J. JUNG, *ibid.* **75** (1992) 2883.
7. L. A. XUE and I. W. CHEN, *ibid.* **76** (1993) 2785.
8. K. P. GADKAREE, *J. Mater. Sci.* **26** (1991) 4845.
9. K. P. GADKAREE and K. CHYUNG, *Am. Ceram. Soc. Bull.* **65** (1986) 370.
10. J. HOMENY, W. L. VAUGHN and M. F. FERBE, *J. Am. Ceram. Soc.* **73** (1990) 394.
11. E. YASUDA, T. AKATSU and Y. TANABE, *J. Ceram. Soc. Jpn.* **99** (1991) 51.
12. M. YANG and R. STEVENS, *J. Mater. Sci.* **26** (1991) 726.
13. T. AKATSU, Y. TANABE and S. MATSUURA *et al.*, *J. Ceram. Soc. Jpn.* **99** (1991) 416.

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