Journal of the European Ceramic Society 20 (2000) 1877–1881

Mechanical properties and fracture behavior of fibrous Al_2O_3/SiC ceramics

Jihong She *, Takahiro Inoue, Masato Suzuki, Satoshi Sodeoka, Kazuo Ueno

Department of Energy Conversion, Osaka National Research Institute, Midorigaoka 1-8-31, Ikeda, Osaka 563-8577, Japan

Received 18 August 1999; received in revised form 8 February 2000; accepted 12 February 2000

Abstract

Fibrous Al_2O_3 ceramics with a mixture of SiC and Al_2O_3 as the cell boundaries were fabricated by extrusion-molding and hot-pressing techniques. The effects of the cell boundary composition on the mechanical properties and fracture behavior are investigated. It is shown that a 65:35 mixture of SiC:Al_2O_3 can act as a suitable cell boundary for Al_2O_3 cells. In bending tests, such a ceramic displays a non-catastrophic fracture behavior with reasonable load-carrying capability, and its fracture energy and apparent toughness are up to 1349 J/m^2 and 6.0 MPa $m^{1/2}$, respectively. © 2000 Elsevier Science Ltd. All rights reserved.

Keywords: Al_2O_3–SiC; Composites; Fibres; Fracture; Mechanical properties

1. Introduction

The incorporation of high-strength ceramic fibers into ceramic matrices has led to a new generation of advanced composites with high toughness and graceful failure characteristics. However, the fabrication of fiber-reinforced ceramic composites is time-consuming, complex, and expensive. Therefore, some simple and inexpensive processes are expected. Coblenz[1] have successfully developed a novel and powerful method to make ceramic composites with a distinct fibrous texture, consisting of high-aspect-ratio polycrystalline cells and thin cell boundaries. This structure can be obtained from ordinary ceramic powders via conventional ceramic- and polymer-processing technology. Specifically, the "green" polymer filaments containing the ceramic powder as the polycrystalline cell are coated with a secondary phase as the cell boundary, then compacted and sintered, resulting in a wood-like fibrous microstructure. The resultant ceramics are called "fibrous ceramics".

Following Coblenz's method, Baskaran et al.[2–4] have fabricated a variety of fibrous ceramics such as SiC/C, SiC/BN and Si_3N_4/BN. Due to the preferred crack propagation along the weak C or BN boundaries, these fibrous ceramics fail non-catastrophically in a similar manner to fiber-reinforced ceramics. However, this graceful failure has not been demonstrated for fibrous Al_2O_3 ceramics, although several systems such as Al_2O_3/ZrO_2, Al_2O_3/Al_2TiO_5 and Al_2O_3/Ni have been investigated.

The objective of the present work is to show that a non-catastrophic fracture behavior can be achieved for fibrous Al_2O_3 ceramics with a mixture of SiC and Al_2O_3 as the cell boundary. The fracture behavior in combination with the mechanical properties such as strength, toughness and fracture energy are evaluated in bending.

2. Experimental procedure

Green Al_2O_3 fibers were prepared using an extrusion process. The starting material was α-Al_2O_3 powder (0.22 μm, TM-D, Taimei Chemicals Co., Ltd., Japan). 17.5 vol% ZrO_2 (0.30 m, TZ-8Y, Toyo Soda Manufacturing Co., Ltd., Japan) was added to develop a fine-grained microstructure. To form green fibers by extrusion, some organic agents such as binder, softener and dispersant must be used. In this work, 4 wt% methyl cellulose and 3.5 wt% dynamite glycerol were employed as binder

---

* Corresponding author present address: Institute of Materials Research, German Aerospace Centre (DLR), 51147 Köln, Germany. Fax: +49-2203-696480.

E-mail address: jhshe@hotmail.com (J. She).

PII: S0955-2219(00)00067-4

and softener, while 2 wt% glycol together with 1.5 wt% polyglycol were used as dispersants.

Al_2O_3 and ZrO_2 powders were mixed with methyl cellulose in acetone by ball milling using alumina grinding media. After drying at room temperature and sieving through a 32-mesh screen, the mixture was further mixed with a "liquid" mixture, which was prepared by mixing dynamite glycerol, glycol and polyglycol in water using magnetic stirrer. The relative amount of water to (Al_2O_3 + ZrO_2) powders was taken as 15 wt% to ensure a relatively homogeneous distribution of ceramic particles and organic components. Extrusion was performed on a laboratory-scale extruder. The diameter of green fibers was fixed at about 0.5 mm.

Sheets of unidirectionally aligned green fibers were produced by winding the fibers around a mandrel and fixing them into place with an adhesive. After trimming to 39.5 mm long and 20.5 mm wide with green fibers aligned in the long direction, these sheets were coated by spraying with $SiC+Al_2O_3$ slurries, which were prepared by milling SiC and Al_2O_3 powders in ethanol. The content of powder solids in the slurries was ~7.5 wt%. The volume ratio of SiC to Al_2O_3 was taken as 50:50, 65:35 and 80:20 to determine the effects of the cell-boundary composition. Fibrous specimens were assembled from the coated fiber sheets. Typically, 18 sheets were stacked in a graphite die of 21× 40 mm, and uniaxially pressed at 250°C using a 30-MPa pressure. This collapsed and consolidated the fibers into a solid billet. Hot-pressing was carried out in vacuum under 25 MPa at 1500°C for 1 h, and a low heating rate of 5°C/min was used below 600°C to remove the organic polymers. After hot-pressing, the billets were cut and ground into rectangular bars of 3 mm (width) × 4 mm (height) × 40 mm (length), with the tensile surface normal to the hot-pressing direction and with the grinding direction parallel to the length of the specimen.

Density was measured by water immersion method. Flexural strength was determined by a three-point bending test with a support distance of 30 mm and a cross-head speed of 0.5 mm/min. The tensile surfaces were polished and the edges were chamfered. Fracture toughness was measured by a four-point SENB technique with a 10-mm inner span and a 30-mm outer span at a cross-head speed of 0.1 mm/min. The notch depth and width were about 1.5 and 0.1 mm, respectively. Strength and toughness measurements were conducted in a universal testing machine with the tensile surface perpendicular to the loading direction. Load and displacement data were recorded with a computerized data- acquisition system. At least three bars were tested for each cell-boundary composition. Fracture energy was calculated from the area under the load–displacement curve of the notched bar. Cross sections and fracture surfaces were observed by optical and scanning electron microscopy, respectively.

3. Results and discussion

Fig. 1 shows the polished cross-sectional surface of a representative specimen, in which the cell and cell boundary structures are clearly visible. The polycrystalline Al_2O_3 cells (gray) are uniformly separated by the continuous SiC cell boundaries (dark). Due to the deformation of the green fibers along the compression axis during warm-pressing and hot-pressing, the Al_2O_3 cells appear as the flattened hexagons with an aspect of ~2. The width of the cells are 540 ± 8 μm. The volume fraction of the cell boundaries, V_{Boundary}, was determined to be about 10% from 300-cell encompassed area on the cross section using optical microscope with an image processor-analyzer. On the other hand, SEM observations revealed that the Al_2O_3 cells were almost free of pores, but a few of pores were presented in the SiC cell boundaries due to the poor sinterability of SiC. During grinding and polishing, the SiC-containing boundaries are easy to be removed, and thus appears as grooves on the polished surfaces, as shown in Fig. 1. Furthermore, the density of the SiC cell boundaries was estimated from

$$\rho_{\mathrm{ceramic}} = \rho_{\mathrm{cell}} \cdot \left(1 - V_{\mathrm{boundary}}\right) + \rho_{\mathrm{boundary}} \cdot V_{\mathrm{boundary}} \qquad (1)$$

where ρ_{ceramic}, ρ_{cell} and ρ_{boundary} are the bulk densities of the fibrous ceramics, the Al_2O_3 cells and the SiC cell boundaries, respectively. In this work, the densities of the fibrous specimens were measured to be 4.1504, 4.0459 and 3.9927 g/cm^3 when the cell boundaries contained 50, 65 and 80 vol% SiC. Table 1 presents the estimated densities of the SiC cell boundaries for specimens with different SiC contents in the boundaries. Also, the relative densities and the total porosities for the SiC cell boundaries are given in Table 1. In calculations, the density of the Al_2O_3 cells was taken as the

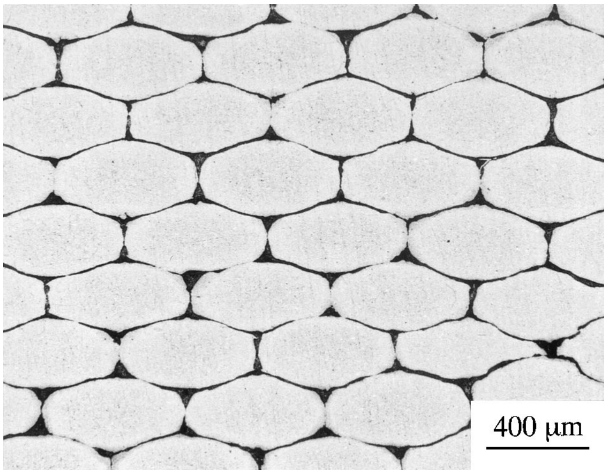

Fig. 1. Optical micrograph of the cross-section of a representative specimen.

Table 1
Density and porosity for the SiC-containing boundaries in fibrous Al_2O_3/SiC ceramics

Boundary composition	Bulk density (g/cm^3)	Relative density (%)	Total porosity (%)
Al_2O_3–50 vol% SiC	2.822	78.7	21.3
Al_2O_3–65 vol% SiC	1.777	51.1	48.9
Al_2O_3–80 vol% SiC	1.245	37.0	63.0

theoretical value of 4.298 g/cm^3, and the theoretical densities of the SiC-containing boundaries were estimated using a rule of mixtures with the values of 3.965 g/cm^3 for Al_2O_3 and 3.21 g/cm^3 for SiC. It can be seen in Table 1 that the total porosities in the 65 and 80 vol% SiC-containing boundaries are up to 48.9 and 63.0%. This may greatly weaken the cell boundaries.

Fig. 2 shows the flexural strength behavior of representative specimens. As shown, all the specimens behave like monolithic ceramics until a maximum flexural stress is reached on the tensile surface. Beyond the stress maximum, the load-bearing ability depends strongly upon the composition of the cell boundaries. When the SiC content in the cell boundaries was 50 vol%, failure occurred in a brittle manner. The average flexural strength of three specimens was 263 MPa. Two specimens with higher strengths broke into two parts; but the third specimen with the lowest strength remained intact, with some load-carrying ability (as shown in Fig. 2). The fracture surface of a broken specimen in Fig. 3(a) shows the stepped fracture at only several boundaries.

With 65 vol% SiC in the cell boundaries, the specimens exhibit a non-catastrophic fracture behavior, with significant load-retaining capability beyond the peak stress. The average flexural strength is 283 MPa, slightly higher than that of the specimens with 50 vol% SiC-containing boundaries. Examination of the specimens after testing showed that fracture originated at the tensile surface between two supporting points, followed by shear delamination almost parallel to the surface along the cell boundaries. This should be attributed to a relatively low shear strength of the 65 vol% SiC-containing boundaries. The terraced topography in Fig. 3(b) indicates the preferential crack growth along the cell boundaries, which appear bright in the SEM micrograph.

Specimens with the cell boundaries containing 80 vol% SiC display a graceful failure, with some load retention at specimen deflections as large as ~1.0 mm. The mean flexural strength was 106 MPa, with strengths ranging from 90 to 131 MPa. The low flexural strength suggests the decrease in the shear strength of the cell boundaries.

Fig. 4 shows the load displacement curves of notched specimens under four-point bending tests. As shown, the fracture behavior of all the notched specimens is non-catastrophic, with the evidence of a gradual load decrease after the peak load. During tests, the shear delaminations were observed to occur along the cell

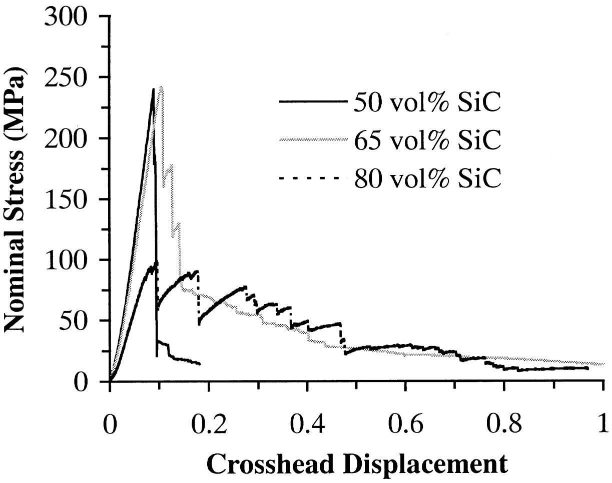

Fig. 2. Nominal stress as a function of crosshead displacement for unnotched specimens with 50, 65 and 80 vol% SiC in the boundaries.

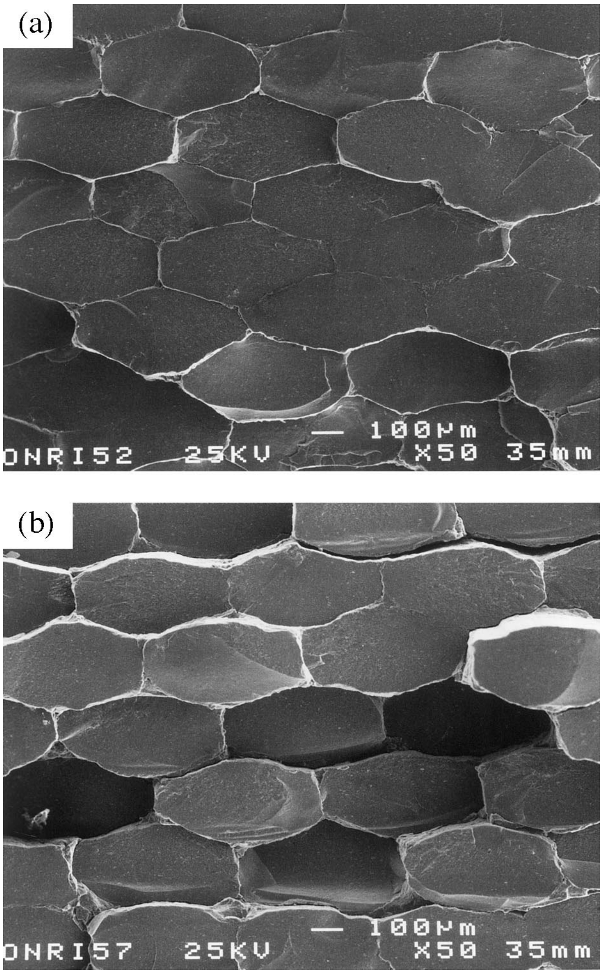

Fig. 3. Fracture surfaces of fibrous Al_2O_3 ceramics with (a) 50 vol% and (b) 65 vol% SiC in the cell boundaries.

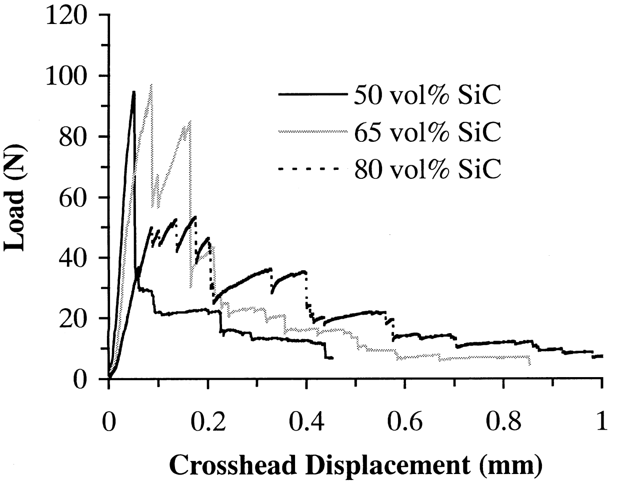

Fig. 4. Load–displacement curves of notched specimens with 50, 65 and 80 vol% SiC in the boundaries.

boundaries, as indicated in Fig. 5(a) by arrows. After tests, all the specimens do not fall apart.

Table 2 summarizes the mechanical properties of fibrous Al_2O_3 ceramics with different volume fractions of SiC in the cell boundaries. When the cell boundaries contain 80 vol% SiC, the measured apparent toughness from the maximum load is only about 2.4 MPa $m^{1/2}$, but the calculated fracture energy from the area under the load displacement curve is up to 1616 J/m^2. In contrast, the specimens with 50 vol% SiC-containing boundaries have a high apparent toughness of 5.1 MPa $m^{1/2}$ but a low fracture energy of 521 J/m^2. These results are considered to be mainly associated with the shear strength of the cell boundaries. Due to a very low shear strength of 80 vol% SiC-containing boundaries, delaminations may occur along almost each successive cell boundary, giving rise to a high energy absorption during the crack extension. When the cell boundaries contain 50 vol% SiC, the shear strength is not low enough for effective delaminations. In this case, the delamination cracks will kink out of the cell boundaries after propagating only a short distance, as shown in Fig. 5(b). This may greatly decrease the energy dissipation capability during fracture. Only in 65 vol% SiC-containing boundaries, the shear strength is at a moderate level. As a result, the specimens exhibit sufficient flexural strength (283 MPa), apparent toughness (6.0 MPa $m^{1/2}$) and fracture energy (1349 J/m^2). These properties are comparable to those of non-oxide systems such as SiC/C and SiC/BN,[3,4] but far superior to those of oxide systems such as Al_2O_3/Al_2TiO_5 and Al_2O_3/Ni.[2,5]

Table 2
Mechanical properties of fibrous Al_2O_3/SiC ceramics with different boundary compositions

Boundary composition	Strength (MPa)	Toughness (MPa $m^{1/2}$)	Fracture energy (J/m^2)
Al_2O_3–50 vol% SiC	263±23	5.1±0.2	521±125
Al_2O_3–65 vol% SiC	283±41	6.0±0.5	1349±109
Al_2O_3–80 vol% SiC	106±16	2.4±0.2	1616±141

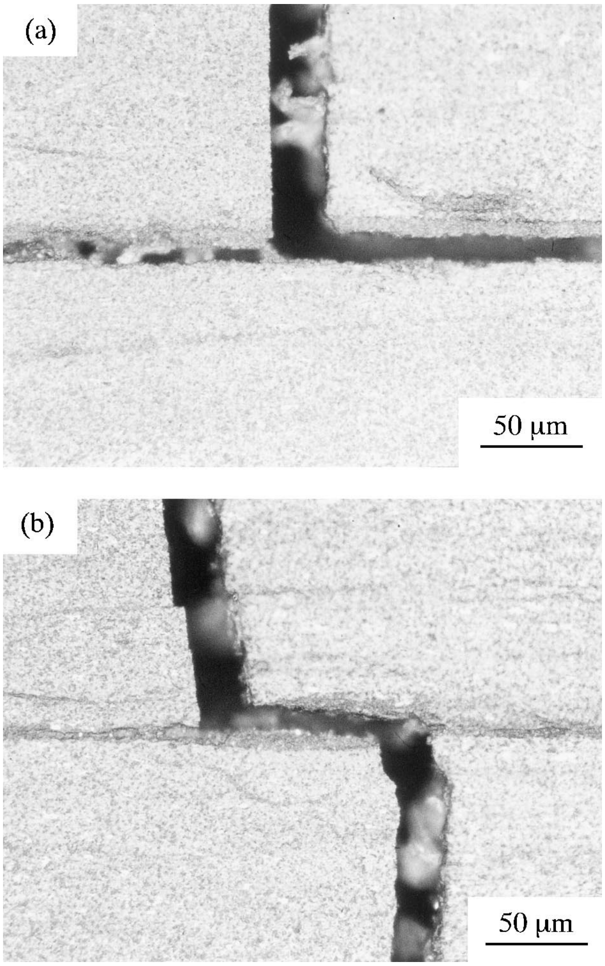

Fig. 5. High-magnification optical micrograph of the side surface of a fibrous Al_2O_3 ceramic with (a) 65 vol% and (b) 50 vol% SiC in the cell boundaries.

4. Conclusions

Fibrous Al_2O_3/SiC ceramics, in which high-aspect-ratio polycrystalline Al_2O_3 cells were separated by thin SiC-containing boundaries, were fabricated by extrusion-molding and hot-pressing. The mechanical properties and fracture behavior were evaluated in bending with a ground or notched tensile surface. When the cell boundaries contained 65 vol% SiC, pronounced shear delamination occurred along the cell boundaries, resulting in a graceful failure. The flexural strength, fracture energy and apparent toughness of such composites were measured to be 283 MPa, 1349 J/m^2 and 6.0 MPa $m^{1/2}$, respectively. These properties have rarely been observed in oxide systems, and are comparable to those of non-oxide systems.

Acknowledgement

Jihong She would like to express his gratitude towards the Agency of Industrial Science and Technology (AIST), Ministry of International Trade and Industry (MITI) for an AIST Research Fellowship.

References

1. Coblenz, W. S., Fibrous monolithic ceramic and method for production. US Patent No. 4772524, 1988.
2. Baskaran, S., Nunn, S. D., Popovic, D. and Halloran, J. W., Fibrous monolithic ceramics: I. Fabrication, microstructure, and indentation behavior. *J. Am. Ceram. Soc.*, 1993, **76**, 2209–2216.
3. Baskaran, S. and Halloran, J. W., Fibrous monolithic ceramics: II. Flexural strength and fracture behavior of the silicon carbide/graphite system. *J. Am. Ceram. Soc.*, 1993, **76**, 2217–2224.
4. Baskaran, S. and Halloran, J. W., Fibrous monolithic ceramics: III. Mechanical properties and oxidation behavior of the silicon carbide/boron nitride system. *J. Am. Ceram. Soc.*, 1994, **77**, 1249–1255.
5. Baskaran, S., Nunn, S. D. and Halloran, J. W., Fibrous monolithic ceramics: IV. Mechanical properties and oxidation behavior of the alumina/nickel system. *J. Am. Ceram. Soc.*, 1994, **77**, 1256–1262.