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In-Situ Characterization of SiC–AlN multiphase ceramics

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AlN and SiC can react and form a solid solution at temperatures above 1800 °C, a result that may be beneficial for sintering silicon carbide ceramics. The pressureless sintered AlN–SiC multiphase ceramics have reached high density at a temperature of 2100 °C for 1 hr in Ar. Analytical scanning transmission electron microscopy was then used to determine the grain boundary, fracture surface, and the local compositions. Because AlN has a higher solid vaporization pressure than SiC, the vaporization rate of the AlN solid would far exceed that of SiC at a sintering temperature. The vaporizing AlN was deposited on the surface of SiC powder; SiC grains then elongated in a random arrangement. The form of elongated rod crystals of 4H SiC is 5 to 8 μm in length and 1 μm in width. It resulted in the sample fracture section producing pulling-out and a strong tearing-open effect. The bending strength and the fracture toughness of the material obtained are 420 MPa and 4.40 MPa $\times$ $m^{1/2}$, respectively. © *1999 Kluwer Academic Publishers*

1. Introduction

Silicon carbide ceramics are of higher hardness, creep, and oxidization resistance, therefore, they are applied for their excellent mechanical properties, particularly at high temperatures. SiC can react with AlN and form an extensive solid solution at temperatures between 1800 to 2100 °C, according to results based on Cutler *et al.* [1]. At present, there is much research on the SiC–AlN system, including the manufacturing of SiC–AlN mixed powders by organo-metallic precursors [2, 3]; carbon-thermal reduction [4, 5]; and the phase diagram of the AlN–SiC system [6–8].

The main purpose of this paper is to study the *in-situ* characterization of enlarging rod crystals of SiC–AlN composition. The dense SiC–AlN multiphase ceramics were fabricated by pressureless sintering.

2. Experimental procedure

SiC–AlN was processed from a mixture of α-SiC, AlN, and Y_2O_3 powders in which Y_2O_3 was a sintering additive. The average size of SiC and AlN particles was 0.6 and 3 μm respectively, which was measured by the conventional powder weight distribution method. Powder of monolithic α-SiC was mixed with AlN and Y_2O_3 by ball-milling for 10 h. The weight ratio of composition the SiC : AlN was 80 : 20. The morphology of powders is shown in Fig. 1. The mixture of powders was formed to bar samples by CIP and sintered in a graphite die. By pressureless sintering at 2100 °C for 1 h in Ar, dense multiphase ceramic materials were attained for a binary composition of SiC–AlN.

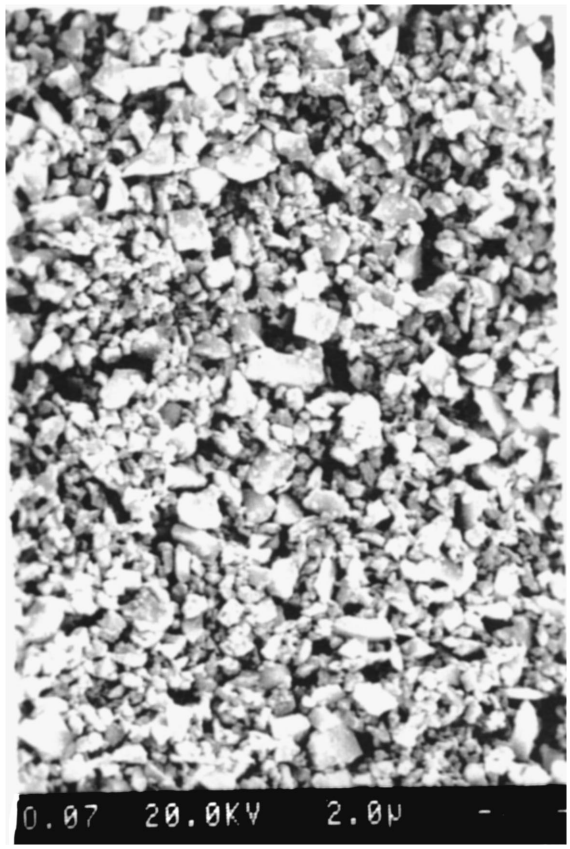
The flexural strength measurements were ground to a dimension of 2.5T $\times$ 5W $\times$ 26L mm^3 from bar specimens of 8T $\times$ 8W $\times$ 30L mm^3. The strength at room temperature was measured by a three-point bending test. The fracture toughness of the materials was evaluated using specimens tested in the direct measurement method.

The microstructure of the boundary interface of the SiC–AlN multiphase ceramics were observed by scanning electron microscopy (SEM) and transmission electron microscopy (TEM) with an energy dispersive X-ray spectrometer (EDXS) for elemental analysis.

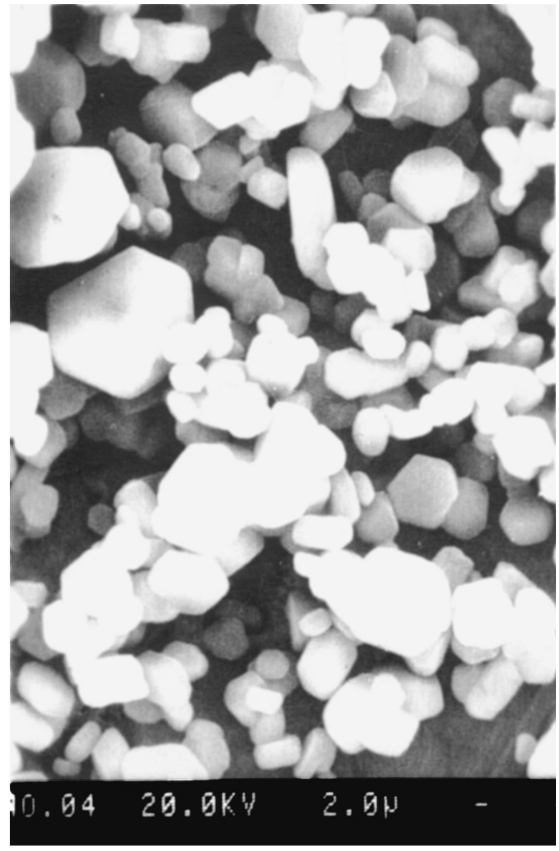
3. Results and discussion

3.1. Mechanical properties

The bending strength at room temperature was measured by a three-point bending test with a specimen size of 2.5T $\times$ 5W $\times$ 26L mm^3. The test was conducted by using the Instron's materials testing machine, and the deformation rate was 0.5 mm/min. The fracture toughness of SiC–AlN multiphase ceramics was evaluated using specimens tested in the indentation fracture toughness. K_{IC} was examined at a load of 10 kg according to the direct-crack measurement method [9]. $K_{IC} = X_r P / C^{3/2}$. The strength and toughness values of SiC–AlN multiphase ceramics are shown in Table I.



(a) SiC



(b) AlN

Figure 1 SEM micrograph of powder shape.

TABLE I The mechanical properties of PLS SiC–AlN multiphase ceramics

SiC : AlN (wt %)	Bending strength (MPa)*	Fracture Toughness (MPa × $m^{1/2}$)**
80 : 20	420	4.40

*: Three-points.
**: $K_{IC} = X_r P / C^{3/2}$

The pronounced fracture surface roughness is shown in detail in Fig. 2. Pulling-out and tearing-open mechanisms may have occurred.

3.2. Microstructure

The microstructure is composed of elongated grains, as shown in Fig. 3. The crystals are randomly distributed. The long axis measures between 5 to 8 μm, and the short axis is approximately 1 μm long. Because they are not present in the starting powders and were formed by a combined reaction and grains growth during sintering. Through observation by TEM with EDXS, the elongated grain core of SiC was identified as a 4H type, as shown in Fig. 4.

3.3. *In-situ* characterization

If two materials have the same crystals structure similar atomic or ionic sizes, and a similar type bonding, the

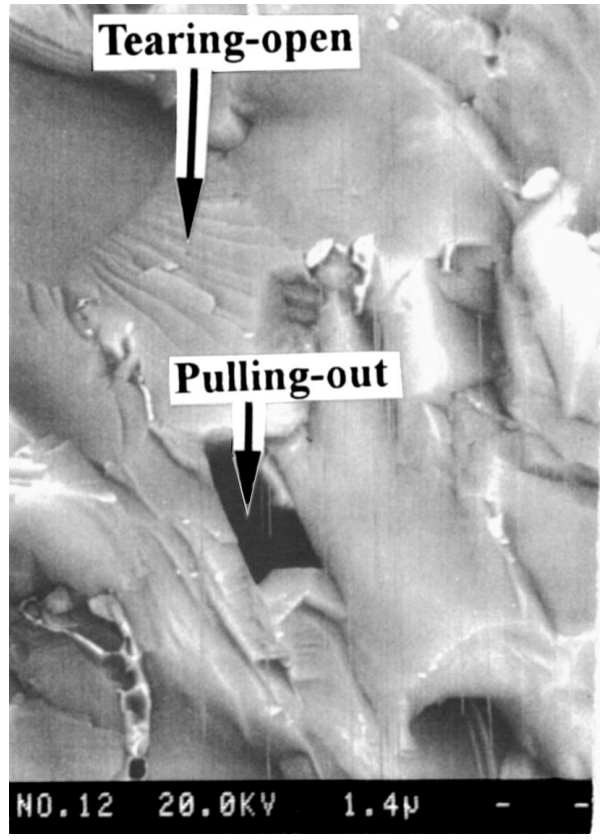


Figure 2 The fracture of SiC–AlN pulling-out and tearing-open phenomenon.

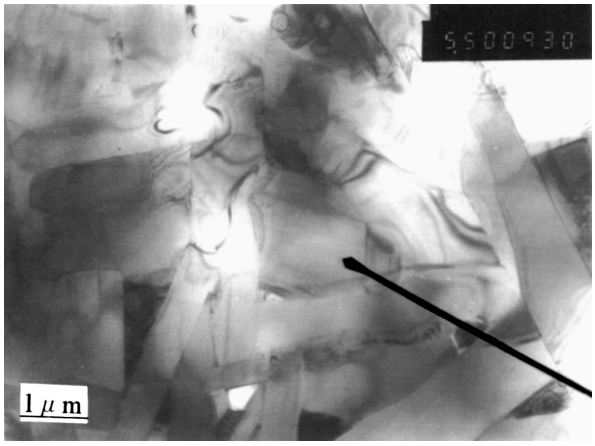
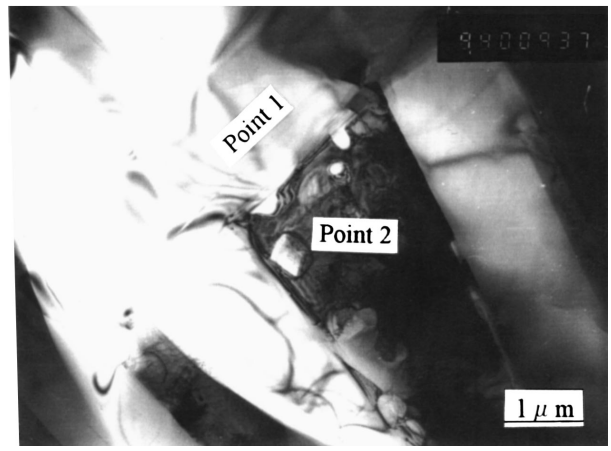


Figure 3 The fracture surface of SiC–AlN showing rods structure.



(a) TEM image of the microstructure of SiC–AlN.

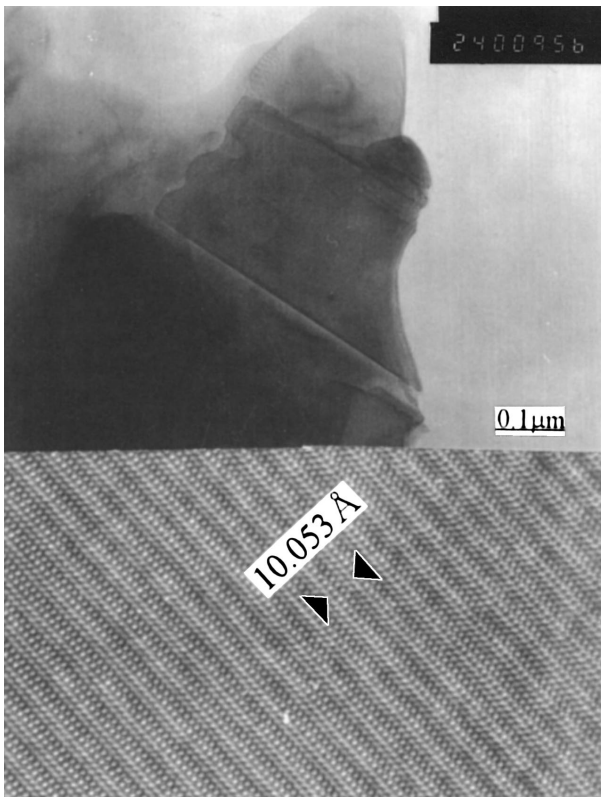
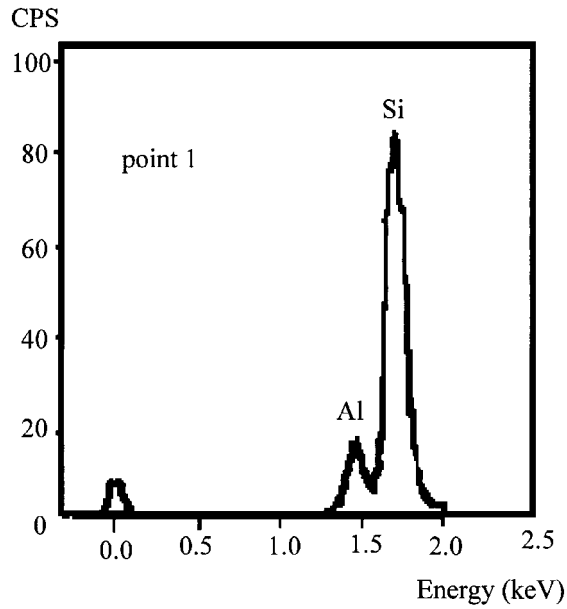
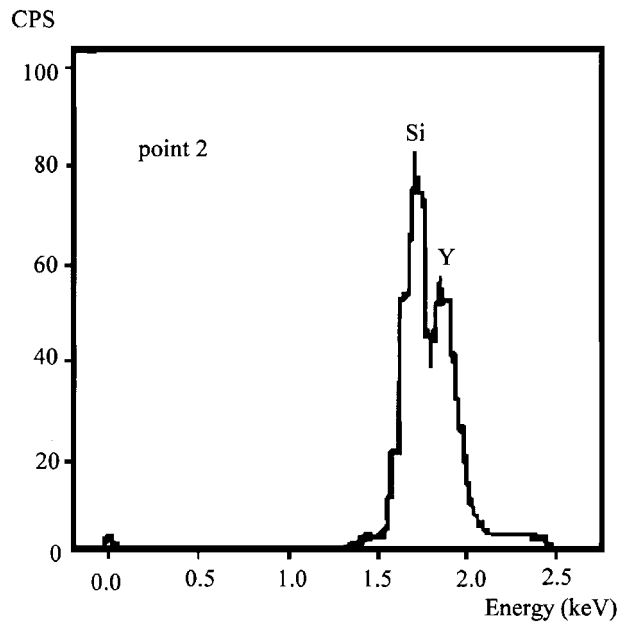


Figure 4 The 4H phase of SiC rod crystal core by TEM.



(b) The EDXS image at the position of point 1 of rod crystal.



(c) The EDXS image at the position of point 2 of triangular area.

Figure 5 The boundary of both the elongation crystals and triangular area.

binary compositions easily form a solid solution. α-SiC is a covalent compound of hexagonal crystal structure, AlN is the same as α-SiC. For the hexagonal structure and lattice parameters, they are very close. Their properties are listed in Table II.

Further observation of the microstructure of SiC–AlN multiphase ceramics using TEM with EDXS, found the difference in composition for both the elongation crystals and triangular area, as shown in Fig. 5.

The solid vaporization pressures of AlN and SiC achieve 14 and 1.23×10^{-3} mmHg at 1900 °C [10]. Of course, the vaporization rate of the AlN solid far exceeds that of SiC at 2100 °C. It may be imagined the change in the microstructure during the sintering process. AlN vaporized the first particles and deposited them on the SiC surface, and they adhere firmly to the surface. AlN vaporized the second particles from its

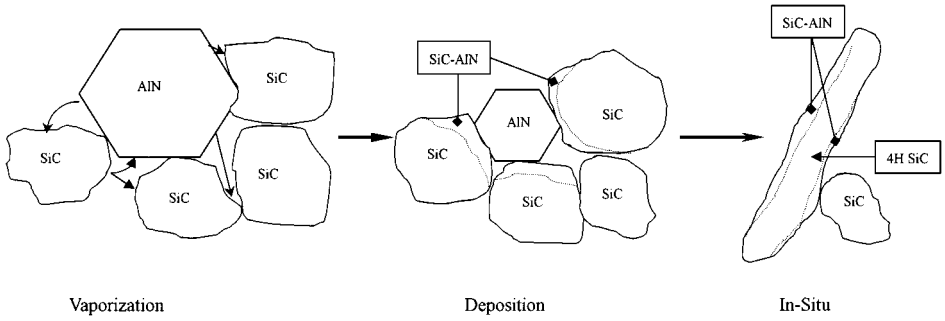


Figure 6 The schematic for in-situ characterization of SiC–AlN multiphase ceramics.

TABLE II Some physical properties of SiC and AlN

Materials and Properties	α-SiC	AlN
Density (g/cm^3)	3.21	3.26
Crystal structure	Hexagonal	Hexagonal
Lattice constant (nm)	$a_0 = 0.30763$, $c_0 = 0.50480$	$a_0 = 0.31114$, $c_0 = 0.49792$
Thermal expansion coefficient ($10^{-6} \cdot K^{-1}$)	4.5	6.09
Young's modulus (GPa)	410	350
Thermal conductivity (w/m $\cdot$ K)	54.43	30.10

large powder surface and decreased their size until completely deposited on the SiC surface. Subsequently, AlN adhered firmly to the SiC surface and diffused into SiC grain body; finally SiC and AlN forms SiC–AlN solid solution layer on the surface of SiC grains. Because the sintering process took only 1 h, the AlN may not completely diffuse into SiC grains and will not form a solid solution with SiC. Thus, the microstructure is not affected to form the homogenous structure of the SiC–AlN solid solution. This result may be proven by TEM, shown in Fig. 5. Point 1 in Fig. 5 indicates the composition of the rod crystal of 4H SiC. Point 2 in Fig. 5 indicates the composition of the triangular area between rod crystals, in which there is no AlN. To consider the surface area of SiC powders, SiC and AlN would form a solid solution where AlN existed. Then, SiC–AlN was a barrier to resist SiC grain enlargement. On the other hand, opposite to the phenomena on the part of SiC surface in which no AlN deposited, SiC grains were free to enlarge during the sintering process. Therefore, SiC grains became rod crystal and were composed of disorder or crisscross microstructure. It may be imagined the change in the microstructure during the sintering process, as schematically shown in Fig. 6.

4. Conclusion

SiC–AlN multiphase ceramics can be fabricated by pressureless sintering of compacted mixed powders at 2100 °C for in an Ar atmosphere. After sintering, SiC grains would elongate rod crystals microstructure in which the arrangement was random. The form of elongated rod crystals of 4H SiC is 5 to 8 μm in length and 1 μm in width.

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