Journal of the European Ceramic Society **19** (1999) 2033–2038

PII: S0955-2219(98)00197-6
0955-2219/99/$ - see front matter

Mechanical Property Improvement and Microstructure Observation of SiCw–AlN Composites

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(Received 20 May 1998; accepted 7 August 1998)

Abstract

SiCw–AlN composites were produced with different sintering additives. Eight wt% Y_2O_3 was found a suitable sintering additive for mechanical property improvement of SiCw-AlN composites. With the sintering additive, SiCw–AlN composites with whisker content ranging from 10–30 wt% were hot-pressed to high density, and the flexural strength and fracture toughness were significantly improved with the increase of SiC whisker content. SEM and TEM observation indicated that several kinds of toughening mechanism contributed to the increasing toughness, including crack deflection, crack branching and whisker bridging process. The interfacial boundaries of the composites were also discussed in detail. © 1999 Elsevier Science Limited. All rights reserved

Keywords: whiskers, mechanical properties, composites, SiC, AlN.

1 Introduction

AlN ceramics attract growing interest for their high thermal conductivity and good electrical properties in recent years. The theoretical thermal conductivity of AlN single-crystal was estimated at $320\,W\,m^{-1}\,K^{-1}$, which is 80% of the thermal conductivity of copper metal. According to the microstructure and chemical composition of the sintered ceramics, thermal conductivity of AlN ceramics varies between 80 and $260\,Wm^{-1}\,K^{-1}$.[2–5] Besides high thermal conductivity, AlN ceramics exhibit good electrical insulation, low dielectric constant and low thermal expansion coefficient. It is commonly recognized that AlN ceramics are considerably potential substrate materials.

In addition, AlN ceramics have various structural and refractory applications. They are also expected to be used as engine components, such as heat exchanger in gas turbine or a matrix for thermal shock resistant ceramic–ceramic composites due to their corrosion resistance and high-temperature stability. However, in many cases, the relatively low flexural strength and fracture toughness of AlN ceramics limit their application in modem industries and high-technologies.[6]

In the past years, a lot of work were done on SiC whisker reinforcement of ceramics. It was found that SiC whisker reinforcement of ceramics could result in substantial improvement in flexural strength, fracture toughness and wear resistance.[7–10] For example, the fracture toughness of Al_2O_3 was doubled or tripled to approximately $9{\cdot}5\,MPa\,m^{1/2}$ with the addition of 30 vol% SiC whiskers.[11,12] The substantial improvement in flexural strength and fracture toughness was achieved by load transfer and other mechanisms, such as crack deflection, crack bowing, whisker bridging and whisker pullout.

Through carefully controlling the processing parameters, interface reaction and microstructure development, incorporating a ductile metal, such as Al, was proved effective in enhancing the mechanical properties of AlN ceramics.[13] Based on the strengthening and toughening mechanism, incorporation of SiC whisker should also be a potential method in improving the mechanical properties of AlN ceramics. AlN has great compatibility with SiC, and a flexural strength of up to 1 GPa was achieved in AlN–SiC material because of the formation of AlN–SiC solid solution with a dense, equiaxed grain structure.[14] However, little work has been done in reinforcing AlN ceramics with SIC whiskers. In this work, we tried to improve the mechanical properties of AlN ceramics by the incorporation of SiC whiskers, and analyze

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the toughening mechanism and interfacial boundaries in the composites through SEM and TEM observation.

2 Experiment Procedure

The starting powders included AlN powders, SiC whiskers, SiO_2 and Y_2O_3 powders. The average grain size of AlN powders was 1 μm, nitrogen content 34·01 wt% and oxygen content 0·60 wt%. SiC whiskers (Advanced Matrix Company) were dimensionally straight with smooth surface. Few clusters were found in the whiskers. The mean length and diameter of SiC whisker were measured as approximately 40 and 0·5 μm, respectively, and the average aspect ratio was 80. They were not coated. Oxygen, carbon and iron were the main impurity. The average particle size of both SiO_2 and Y_2O_3 powders was 1 μm, and the purity was 99·99%.

First, the SiC whiskers were dispersed by ultrasonication and a high-shear homogenizer. Then, the dispersed SiC whiskers, AlN powders and sintering additives of Y_2O_3 and /or SiO_2 were poured together and ball-milled for 12 h in alcohol using Si_3N_4 pot with Si_3N_4 milling balls. The chemical composition of the composites was listed in Table 1. The SiC whisker content in the mixture was 0, 10, 20 and 30 wt%, respectively. The mixture were dried, sieved through a 100 mesh sieve. Then, the mixed powders were compacted into pellets under unidirectional pressure. The pellets were put into BN coated graphite die and hot-pressed in nitrogen gas at 1850°C for 1 h under 20 MPa pressure. The heating and cooling rates were both 10°C $min.^{-1}$

A three-point bending test was used to measure the room temperature flexural strength, with the bar dimensions of 2·5×5×30 mm and the cross-head speed of 0·5 mm min^{-1}. The fracture toughness was measured by the single edge notch beam (SENB), with a center notch 2·0 mm deep in a test bar of 5×2·5×30 mm and the cross-head speed of 0·05 mm min^{-1}. For each batch, five bars were tested and the average values was reported. The density was measured by the Archimedes displacement method. X-ray diffraction was used to identify the phases present on the composites. Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) were used for microstructure analysis. In order to study the whisker/matrix interface, high-resolution electron microscopy (HRFM) was also used. Thin foils of the composites were prepared by dimpling and subsequent ion-beam thinning.

3 Results and Discussion

3.1 Selecting sintering additive for SiCw–AlN composites

In order to study the effect of sintering additive composition on the mechanical properties of SiCw–AlN composites, 20 wt% SiCw–AlN composites with different sintering additive were produced. Table 1 lists the additive composition, relative density, flexural strength and phase composition of 20 wt% SiCw–AlN composites. X-ray diffraction analysis shows that the main phase in the composites was all SiC and AlN, but the secondary phase was controlled by the additive composition. It was reported that solid solution between SiC and AlN was formed in a wide composition range because both SiC and AlN had the similar wurtzite (2H) structure.[14] In present work, however, no solid solution was found on the diffraction patterns. The reason was probably the low sintering temperature, which caused the atoms diffuse lower than that in others' work.[14] During sintering, sintering additives reacted with Al_2O_3 on the surface of AlN to form liquid, enhancing densification behavior, therefore, the secondary phases were controlled by the additives. When Y_2O_3 content was high in the sintering additive, YAM phase formed easily, however, when SiO_2 content was high, 27R sialon formed.

From Table 1, it was clearly seen that the composite obtained high relative density and high flexural strength with the addition of only Y_2O_3 if the same amount of additives was added. When 10 wt% Y_2O_3 was added, the relative density of the composites was 99·0%, and the flexural strength

Table 1. Chemical composition, properties and phase composition of 20 wt% SiCw–AlN composites

Sample	*Chemical compositions*				*Properties*		*Phase composition*	
	AlN (wt%)	*SiCw (wt%)*	*Y_2O_3 (wt%)*	*SiO_2 (wt%)*	*Density (%)*	*Strength (MPa)*	*Main phase*	*Minor phase*
1	70	20	0	10	82	271(±37)	AlN,SiC	27R sialon
2	70	20	5	5	95	380(±71)	AlN,SiC	YAG,27R sialon
3	70	20	10	0	99	508(±59)	AlN,SiC	YAM
4	72	20	8	0	99	520(±67)	AlN,SiC	YAM
5	74	20	6	0	97	480(±53)	AlN,SiC	YAM
6	76	20	4	0	83	330(±48)	AlN,SiC	YAM,YAG

508 MPa. However, when 10 wt% SiO_2 was added, the relative density was 82% only, and flexural strength 271 MPa. It was related with the role of SiO_2 in the sintering. When increasing the amount Of SiO_2, 27R sialon formed. As we know, 27R sialon has elongated shape, similar to SiC whisker, which inhibited the densification. In addition, formation of 27R sialon absorbed liquid, which also reduced the densification.

Y_2O_3 was often used as sintering additive in the sintering of AlN ceramics, and usually 2–3 wt% was enough. But in SiCw–AlN composites, additive should be more than 6 wt% because of the whisker bridging which made the densification difficult. The flexural strength reached the maximum 520 MPa when 8 wt% Y_2O_3 added. Further increasing the additive content decreased the flexural strength. The reason was that strong interface between AlN matrix and SiC whisker formed when too much Y_2O_3 was added, and the formed strong interface limited the strengthening and toughening effect of SiC whisker in the composites.

3.2 Mechanical properties of SiCw–AlN composites

Based on the above study, 8 wt% Y_2O_3 was used as sintering additive to fabricate SiCw–AlN composites with SiC whisker content ranging from 0 to 30 wt%. The mechanical properties of monolithic AlN and SiCw–AlN composites are shown in Fig. 1 The density of all the materials were larger than 98% of theoretic density. The mechanical properties of the composites increased with the increase of SiC whisker content. The flexural strength and fracture toughness increased from 308 MPa, 3·12 MPa $m^{1/2}$ for monolithic AlN to 646 MPa, 7·17 MPa $m^{1/2}$ for 30 wt% SiCw–AlN composites, respectively. The improvement in the mechanical properties of the composites was achieved by the incorporation of strong whiskers. The SiC whiskers compacted in the matrix of AlN was supposed to highly sustain the load and strengthen the grain boundary.

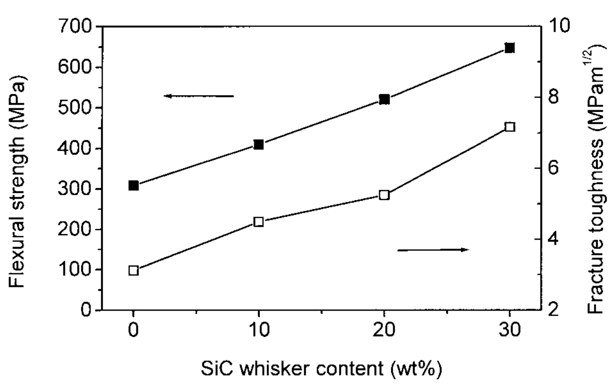

Fig. 1. Mechanical properties of SiCw–AlN composites as a function of SiC whisker content.

3.3 Microstructure observation

Fracture surfaces of monolithic AlN ceramics and 20 wt% SiCw–AlN composites are shown in Fig. 2 The monolithic AlN ceramics were consisted of well crystallized hexagonal AlN grains. It was also revealed that some AlN grains were pulled out from the matrix, indicating the weak bonding of grain boundaries in monolithic AlN ceramics, which resulted in low flexural strength. The intergranular fracture was considered as the main fracture mode. In SiCw–AlN composite, SiC whiskers were almost uniformly distributed within the matrix, orienting with the long axes in planes perpendicular to the hot-pressing direction. The composites were free of significant porosity, although voids associated with whisker agglomerates were occasionally observed. The fracture surface of the composites were quite rough. Crack propagating around the whiskers and whisker pulling out indicated the whisker strengthening effect. It exhibited regular intergranular fracture. The fracture origin areas were located close to the tension surface. However, SEM observation did not reveal the obvious flaw or defects.

Because the elastic modulus and hardness of SiC whisker significantly exceed that of AlN matrix, SiC whiskers were effective in inhibiting the grain

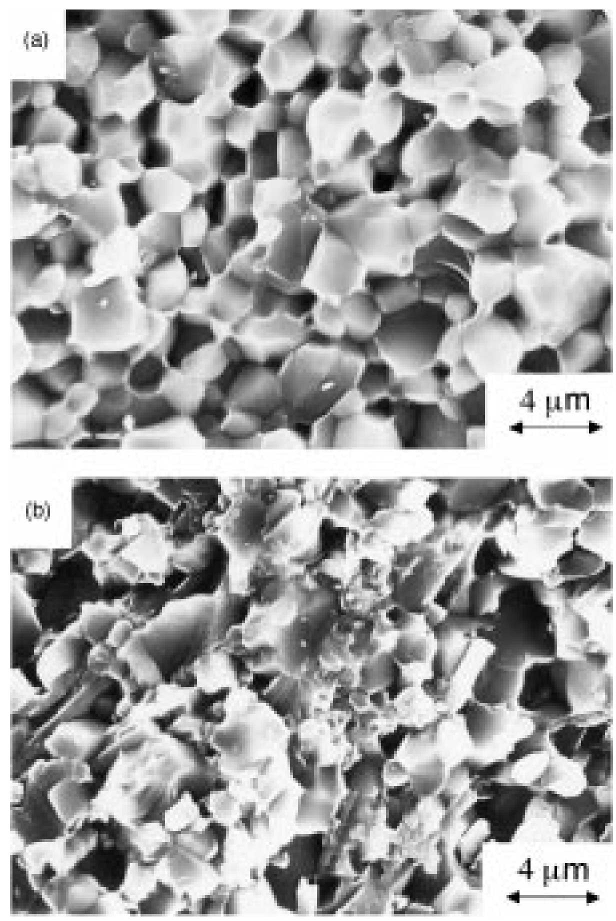

Fig. 2. Fracture surface of (a) monolithic AlN and (b) 20 wt% SiCw–AlN composites.

growth of the matrix and some whiskers were embedded into AlN grains. The grain size of the monolithic was evidently larger than that of the SiCw–AlN composites. Matrix grain refinement was considered one of the reason for the improved flexural strength. In SiC–AlN solid solution system, mechanical properties were greatly improved because of the grain refinement. Flexural strength of 1 GPa was obtained for 75SiC–25AlN composites which contained a dense, equiaxed grain of 1 μm.

The following toughening mechanism were revealed in the microstructure observation of the composites: cracking deflection, debonding, bridging, cracking branching and whisker pull out (Fig. 3). Two possible approaches existed when the crack tip reached the whisker. First, crack deflection occurred due to the mismatch in the elastic modulus between SiC whisker and AlN matrix. The deflection length of the crack mainly depended on the radius, length and strength of the whisker. If the high stress on the crack tip was larger than the strength of the whisker, it broke the whisker and thus resulted in crack branching. Such a process also released the high stress concentrated at the crack tip and inhibited crack propagation in the matrix. The second approach was interfacial debonding. when the stress at the crack tip was not high enough to break the whisker, and the interfacial fracture energy was much lower than that of the matrix on the whisker, the crack would propagate along the interface and cause interface debonding. The observation also indicated that the development of interface debonding allowed the matrix crack to proceed without fracturing the whisker and the whisker would bridge the crack behind the crack tip. The contribution of whisker bridging and interface debonding to toughness were discussed in detail by Becher *et al.*[15] It was found that both debonding and bridging would release the stress concentration at the crack tip and inhibit crack propagation. The toughening effect increased with increasing the strength, volume fracture and radius of whiskers.

3.4 Interfacial behavior

For ceramic composites, the nature of whisker–matrix interface is clearly a key factor of the toughness-related property. In the SIC–AlN system, as mentioned above, a solid solution forms over a wide composition range. Although no solid-solution phase was found by XRD analysis in SiCw–AlN composites, the whisker/matrix interaction was detected by high-resolution electron microscope (HREM).

Two kinds of interfacial layer between matrix and whiskers were found in the composites. Most

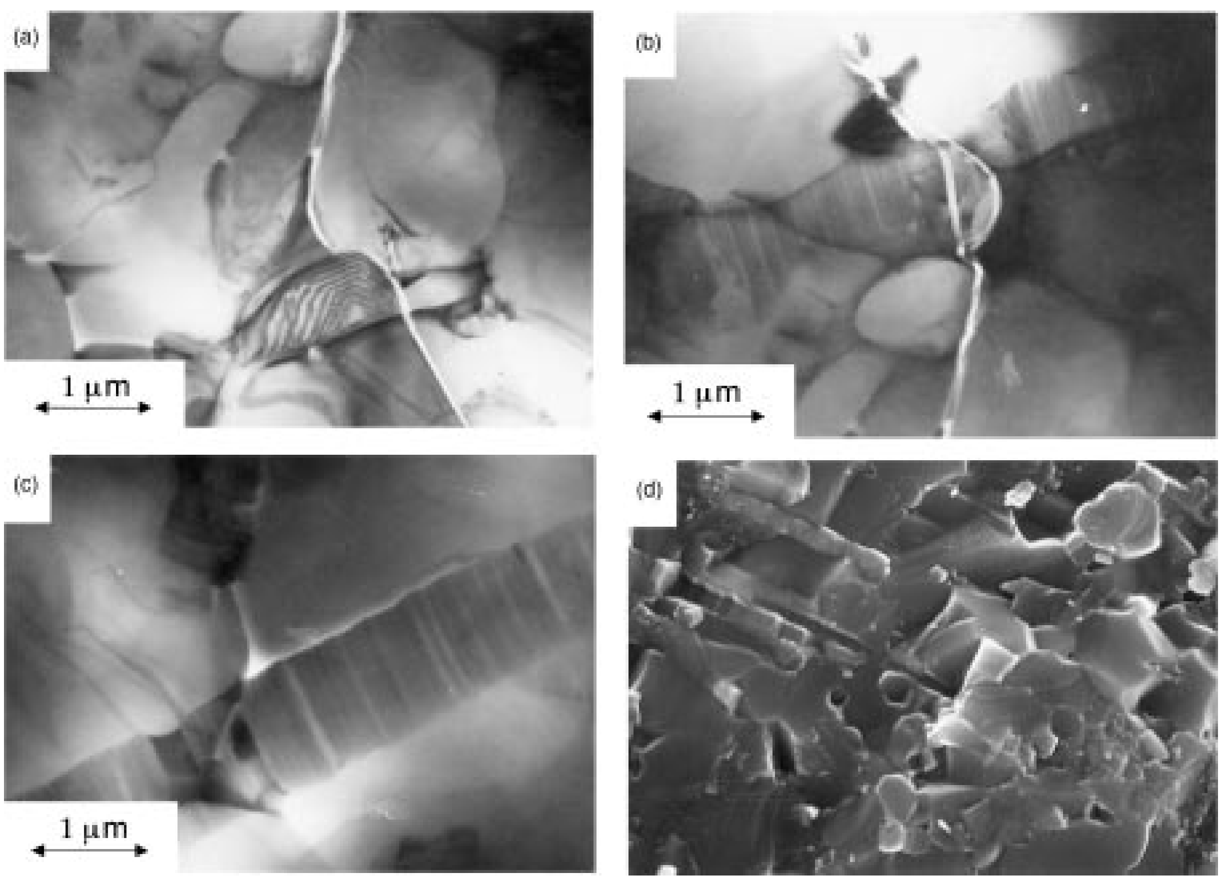

Fig. 3. Toughening mechanisms shown in microstructure observation (a) crack deflection, (b) crack branching, (c) interface debonding, and (d) whisker pullout.

of the interfacial layers were amorphous layers. Figure 4 is a typical HREM image of grain-whisker amorphous interfacial layer with the width of 10A. The HRFM image revealed that no interfacial reaction occurred. A more extensive examination of some of the SiC/AlN interfaces was conducted using analytical electron microscopy fitted with EDAX. EDAX microanalysis revealed the existence of Y in the interfacial layer. The interfacial thin layer was supposed to be a glass phase formed by sintering aids. On the other hand, a few crystallized layers were still found in the composites, as is shown in Fig. 5. EDAX microanalysis revealed the absence of yttrium in the crystallized layers. The crystallized interfacial reaction layers seemed to be the products of solid solution between SiC whisker and AlN matrix.

It is confirmed that interfacial debonding is a prerequisite for effective whisker toughening. In SiCw–AlN composites, when the interface was bonded by an amorphous layer, the weak interfacial might be more likely to debond under stress than a strong interface between SiC whisker and AlN grains. When the interface was bonded by a reaction layer, the whisker would be degraded and its contribution to toughening would be decreased. Figure 6 shows an unusual crack propagation in which the crack proceeded along the whisker growth axis instead of the interface bonding. It also illustrated that the intensity stress of crack tip caused stress stripes in the adjacent grains, but the interfacial boundary was undamaged. One possible explanation of this process was that some solid solution formed over the interface. Due to high strength of SIC–AlN solid solution, this kind of interface had much stronger bonding strength than conventional amorphous interface and decreased the whisker toughening effect.

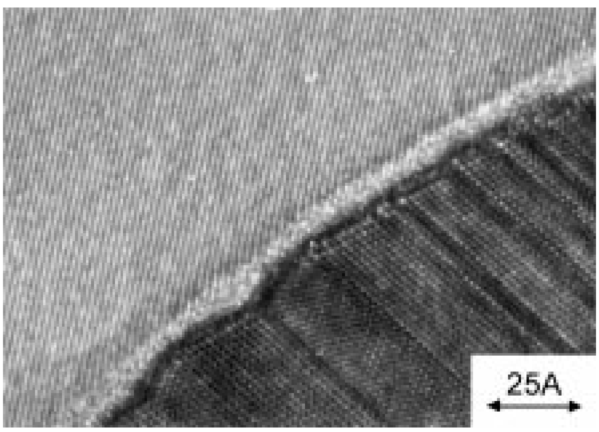

Fig. 4. HREM shows the amorphous interface between SiC whisker and AlN matrix.

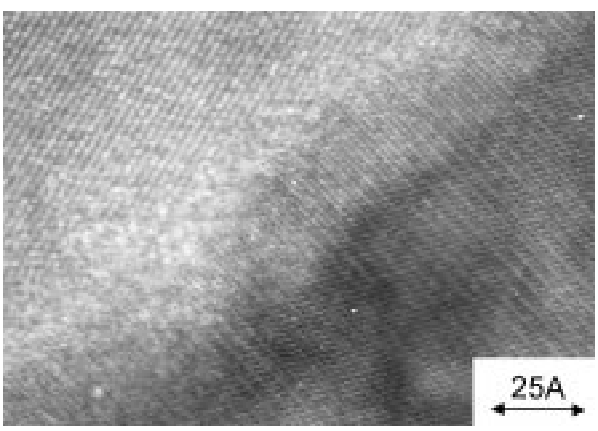

Fig. 5. HREM shows the crystallized interface between SiC whisker and AlN matrix.

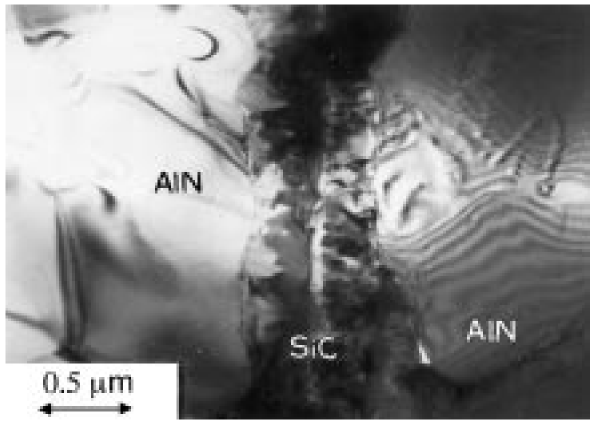

Fig. 6. TEM shows an unusual crack propagation along the whisker growth axis instead of the interface bonding.

4 Conclusions

1. SiCw–AlN composites were fabricated with different sintering additives. It was found that the mechanical properties of the composites reached a maximum when Using 8 wt% Y_2O_3 as sintering additive. It was also indicated that the mechanical properties increased significantly with the increase of SIC whisker content.
2. Microstructure observation of SiCw–AlN composite indicated SiC whiskers were well dispersed in AlN matrix. The main toughening mechanisms revealed by microstructure observation were crack deflection, whisker bridging and interface debonding.
3. Two kinds of matrix/whisker interfaces were found, one was amorphous interface, and the other was crystallized interface. The formation of the crystallized interface depended on processing parameters, and it decreased the whisker toughening effect.

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