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Mechanical Properties of Silicon Nitride–SiC Platelet Composites

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Abstract

Silicon nitride and SiC platelet–silicon nitride composites have been prepared by hot-pressing and by pressureless sintering. As-received, large SiC platelets behave as critical flaws and were separated into two narrow size fractions for the production of composites.

The mechanical behaviour of the composites has been assessed and results are discussed taking into account the raw materials' properties, the microstructure of the dense sintered ceramics and fracture mechanisms.

Si_3N_4- und SiC-Plättchen/Si_3N_4-Keramik wurde durch Heißpressen und druckloses Sintern hergestellt. Unbehandelte, große SiC-Plättchen des Rohmaterials wirken sich wie Mikrorisse aus. Zur Herstellung der Verbundwerkstoffe wurden sie in zwei Fraktionen mit jeweils enger Korngrößenverteilung getrennt. Die mechanischen Eigenschaften dieser Verbundwerkstoffe wurden untersucht und die Ergebnisse werden im Hinblick auf die Eigenschaften der Rohmaterialien, das Gefüge der dichtgesinterten Keramik und die Bruchmechanismen untersucht.

On a préparé du nitrure de silicium et des composites à matrice nitrure de silicium renforcée par des plaquettes de SiC par frittage sous charge et frittage naturel. Les plaquettes de SiC non traitées agissaient comme défauts critiques et ont été séparées en deux fractions de tailles proches pour l'élaboration des composites. Le comportement mécanique des composites a été étudié et les résultats discutés en tenant compte des propriétés des matériaux de départ, de la microstructure des céramiques frittées denses et des mécanismes de rupture.

1 Introduction

Silicon nitride ceramic materials are well known for their excellent high-temperature mechanical properties and stability, and thus can be considered as candidates for high-temperature structural applications. Much effort has been expended in improving the high-temperature strength of these materials by the fabrication of high-purity silicon nitride starting powders showing improved crystallographic and morphological characteristics[1] and also by the development of improved densification additive systems.[2]

Incorporation of secondary reinforcing phases such as particles, whiskers, platelets or short fibres into the matrix can lead to an additional improvement of the silicon nitride matrix toughness, improving reliability. A comparative study of the synthesis and characterisation of silicon nitride–silicon carbide platelet composites (Si_3N_4–SiC(pl))[3] has shown the advantages of using platelets as a secondary phase. Indeed, both the densification inhibition due to the presence of the secondary SiC phase and interactions with secondary liquid phases during sintering, are very limited, thus permitting attainment of improved mechanical properties.

Careful attention has to be paid on the size of the platelets.[4,5] The first commercial SiC platelet

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(SiC(pl)) grades, produced by a North American company (American Matrix) proved to be relatively coarse, up to 70 μm in dimension; such a size can be considered to be larger than the critical defect size for a structural ceramic such as silicon nitride. It is therefore evident that platelet sizes have to be reduced before fabrication of the Si_3N_4–SiC(pl) composites, in order to prevent the generation of supplementary defects.

2 Experimental

The densification characteristics of different commercially available silicon nitride powders during pressureless sintering or hot pressing using 10 wt% Y_2O_3–MgO (1:1 molar proportion) and 1·5 wt% Al_2O_3 + 5·5 wt% Y_2O_3 standard additive systems have been compared previously.[1] Two silicon nitride powders TS10 (Tosoh, Japan) and LC12SX (H. C. Starck, Germany) were selected, on the basis of this comparative study.

The main characteristics of these powders are shown in Table 1. Specific surface area was measured by the BET method; the alpha- and beta-Si_3N_4 content was determined by X-ray diffraction; mean particle size was deduced from Stokes' sedimentation data (Sedigraph, Micromeritics, USA), after ultrasonic dispersion in water using a surfactant. As-received batches of platelets were separated by elutriation–centrifugation in air (Bahco, Sweden) to produce 'fine' and 'coarse' grades. Statistical analysis from scanning electron microscope (SEM) observations showed that the finer platelet grade was of size below 15 μm, with an average dimension of 4 μm, while the coarser grade had a mean size of 9 μm, and a largest size of 30 μm, measured perpendicular to the narrowest dimension of the platelet.

The mixing of the SiC platelets with matrix and additive powders (1·5 wt% alumina + 5·5 wt% yttria), compaction and/or sintering properties and microstructural and X-ray diffraction characterisation of sintered composites have been described elsewhere.[4]

Mechanical properties were assessed on pressureless sintered TS10 matrix composites and hot-pressed LC12SX matrix composites.

Bend strength (σ_F) was measured at temperatures ranging from room temperature to 1300°C, using three-point loading with a span of 15 mm and a crosshead speed of 0·1 mm/min. Strength of hot-pressed materials was measured in a direction parallel to the hot-pressing axis. Critical stress intensity factors (K_{IC}) were measured using the single-edge notched beam technique with a notch width of 190 μm. Room-temperature data were generally obtained from five specimens whereas high-temperature tests were carried out on three specimens.

Table 1. Main powder characteristics of the selected silicon nitride powders

Grade	*Source*	*Oxygen content (mass%)*	*Mean apparent particle size (μm)*	S_S (m^2g^{-1})	β-Si_3N_4 (%)
TS10	$SiCl_4$	1·70	0·4	14	5
LC12SX	Si	1·98	0·4	22	8

3 Results and Discussion

3.1 Mechanical properties of conventionally sintered composites

The room-temperature toughness of the TS10 silicon nitride matrix is improved by incorporation of 10 vol% of either fine or coarse platelets (Fig. 1). Improvement is less substantial using as-received platelets or when introducing higher levels of secondary reinforcing phase, both of which lead to a lower final densification level and, consequently, to a higher residual porosity, limiting the toughness increase.

Microstructural analysis by SEM of fracture faces and of the crack path induced by indentation fracture shows crack deflection around platelets and more probably platelet pull-out during fracture (Fig. 2), explaining the toughness improvement. Both reinforcement mechanisms are a consequence of the very limited interaction between matrix and platelets

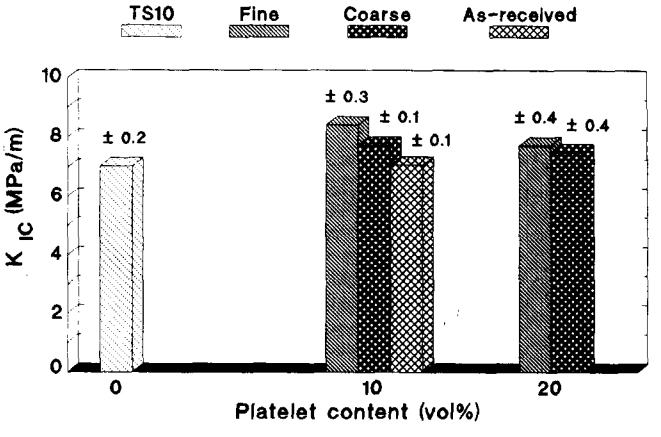

Fig. 1. Room-temperature toughness of pressureless sintered materials.

Fig. 2. SEM micrograph of a fracture face of a platelet composite showing intergranular crack propagation with respect to platelets.

during sintering, leading to weak interfacial bonding betweeen them.[3]

Strength increases slightly only in the case of composites with 10 vol.% of fine platelets (from 612 ± 87 to 671 ± 51 MPa) (Fig. 3). With 10 vol.% of coarse grade, no significant difference is seen, whereas with the as-received total fraction, as well as with 20 vol.% platelets, the strength decreases significantly. Fracture face analysis showed the appearance of coarse platelets at the fracture origin, indicating that platelets induce flaw formation and are likely to be the critical defect, explaining the strength decreases seen with increasing platelet size.

Further decrease of strength with increasing platelet concentration is attributed to the increased porosity resulting from reduced densification.

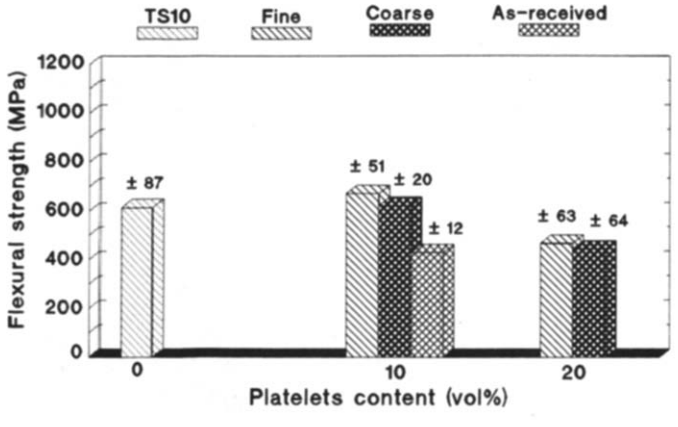

Fig. 3. Room-temperature bend strength of pressureless sintered silicon nitride platelet composites.

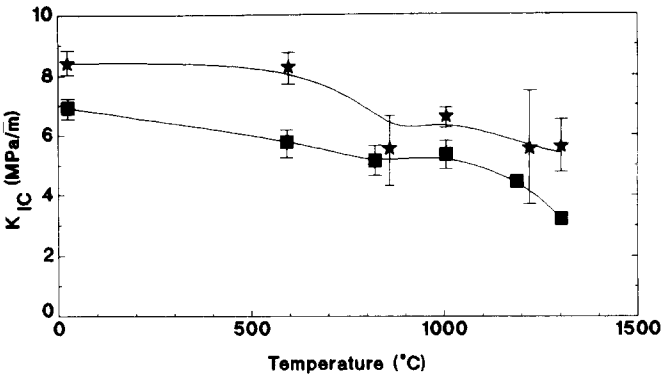

Fig. 4. Temperature dependance of toughness of pressureless conventionally sintered platelet composites (10% 'fine'). ■, K_{IC} matrix; ★, composite.

Indeed, sintered matrix density is 95% of theoretical density, while for composites containing 10 and 20% of platelets, it reduces to 92 and 91% of theoretical density, respectively.

Following these considerations, a composite consisting of a TS10 silicon nitride matrix reinforced with 10 vol.% of fine platelets was fabricated for evaluation of high-temperature mechanical properties. From room temperature up to 1300°C, the toughness of the platelet composite remained higher than that of the silicon nitride matrix material (Fig. 4), while the bend strength was close to that of the matrix in the whole temperature range (Fig. 5). As a consequence, the presence of a larger critical flaw size was assumed and explained mainly by the lower extent of densification of the composite. Above 800°C, toughness and bend strength of both matrix and composite decreased with increasing temperature.

From SEM analysis of the fracture faces, it can be seen that the mode of rupture of the matrix is mixed at 800°C and 1000°C (both inter- and intragranular), and become completely intergranular at 1200°C.

From these observations it is clear that the decrease of the mechanical properties of the matrix material and composite as a function of temperature

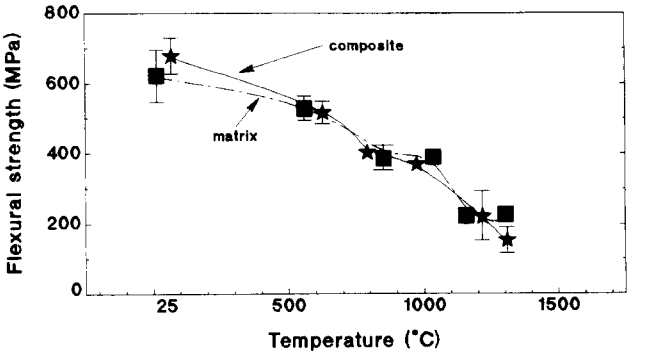

Fig. 5. Temperature dependance of flexural strength of conventionally sintered platelet composite. ■, Matrix; ★, composite.

can be attributed to the degradation of the matrix, and thus to the progressive weakening of the intergranular oxynitride (glass) phase above its glass transition temperature.[6]

The comparison of fracture faces of the silicon nitride material and the composite shows that, up to 1200°C, the fracture face of the composite is rougher than that of the silicon nitride, which may be explained by the occurrence of crack deflection at high temperature, caused by platelets. At 1300°C, the materials were no longer elastic and behaved as they would during creep tests, that is, the interlinkage of cavities probably induced failure (Fig. 6). At this temperature, oxidation of the fracture surface occurred, making SEM analysis at high magnification difficult.

3.2 Mechanical properties of hot-pressed composites

The mechanical properties of hot-pressed composites made from an LC12SX matrix and containing 10 vol.% of fine platelets were evaluated and compared with those of the matrix material. Although mechanical properties as a function of temperature were measured in a direction parallel to the hot-pressing direction, it should be noted that at room temperature identical values were obtained by bending in a perpendicular direction, contrary to what is observed with whiskers,[7] and with in-situ formed elongated β-Si_3N_4 grains.

As was seen in the case of pressureless sintered composites, toughness improvement by platelets occurred from room temperature up to 1300°C (Fig. 7); however, no decrease in K_{IC} was observed with increasing temperature in the hot-pressed samples. Again, evidence of crack deflection appeared from observation of fracture faces which appeared to be much more rough in the case of the composites.

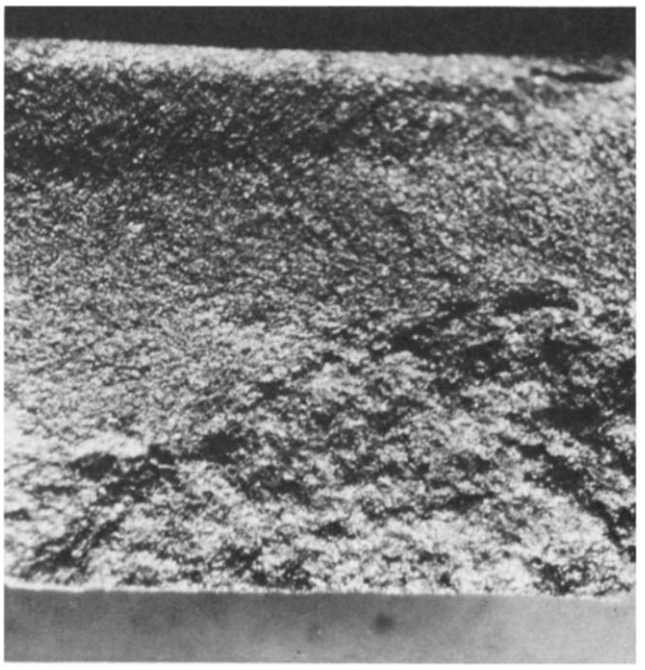

Fig. 6. Fracture face showing creep-like failure of a composite at 1300°C.

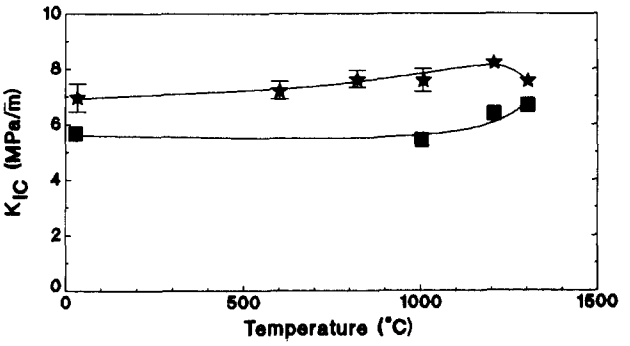

Fig. 7. Temperature dependance of toughness of hot-pressed platelet composites. ■, K_{IC} matrix; ★, K_{IC} composite.

Bend strength was more temperature dependent, as shown in Fig. 8. At room temperature, the strength of the matrix material was higher than that of the composite (1004 MPa compared with 797 MPa for the composite) in spite of the higher toughness of the composite. Therefore, the presence of a larger critical flaw size can be deduced in the case of the composite. Above 800°C, a significant decrease of bend strength was observed for the matrix material, in spite of the fact that its K_{IC} remained almost constant over the whole temperature range; however, the decrease in bend strength of the composite was very limited up to 1200°C. This difference in mechanical behaviour between the matrix material and the composite might be attributed to a higher subcritical crack growth rate in the case of the matrix.[8] This is supported by the observation of a more extended flat area at the fracture origin in the case of the matrix material.

The fracture mode of the matrix observed using SEM was transgranular with respect to the matrix from room temperature up to 1000°C. Above this temperature, surface oxidation makes SEM analysis impossible.

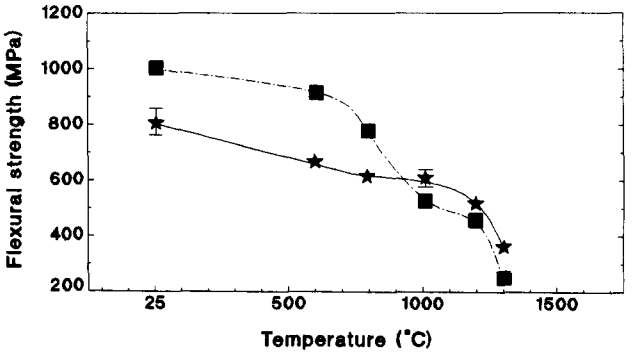

Fig. 8. Temperature dependance of bend strength of hot-pressed platelet composites. ■, Matrix; ★, composite.

At 1300°C, creep-like rupture was again observed, leading to a subsequent decrease of flexural strength of the composite.

4 Conclusions

(a) The toughness of silicon nitride–SiC(pl) composites with 10 vol.% of fine or coarse platelets is greater than that of the matrix material from room temperature up to 1300°C.

(b) At all temperatures crack deflection (or even pull-out) is observed; this is believed to be responsible for the improved toughness.

(c) Simultaneous increase of bend strength is dependent on the platelet size and content. Coarse platelets can be at the origin of the defect.

(d) For hot-pressed materials having a small critical flaw size the bend strength of the composite, even when using fine sized platelets, is less than that of the matrix. However, the strength of the matrix material decreases above 600°C, while that of the composite remains almost unchanged, and above 1000°C, the strength of the composite is higher, attaining 520 MPa at 1200°C.

Acknowledgements

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