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Anisotropic Properties in Hot Pressed Silicon Nitride—Silicon Carbide Platelet Reinforced Composites

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Abstract

The anisotropic properties (microstructure, mechanical properties) of a hot-pressed platelet reinforced silicon nitride composite were compared with those of the monolithic material. The platelets appeared to be orientated with their basal plane in the compressive plane, and to be embedded in a silicon nitride matrix consisting of interlocked elongated β-Si_3N_4 grains with their c axis orientated in this plane. TEM analysis showed an interface, consisting of glassy phase and graphite at the platelet–matrix grain boundary. Moreover the interfacial tensile stresses are in favour of a crack deflection mechanism. It was shown by TEM analysis that crack deflection occurs not only at the silicon nitride–platelet interface, but also at silicon nitride–silicon nitride grain boundaries. The efficiency of this reinforcing mechanism is highly orientation dependent. Because of their two dimensional geometry compared to the one-dimensional β-Si_3N_4 grains, platelets increase the toughness in two dimensions.

Keywords: anistropy, composites, mechanical properties, SiC, Si_3N_4

1 Introduction

Ceramic materials can be reinforced by the addition of secondary phases such as particles, 'short' fibres, whiskers or platelets. The reinforcing mechanism will depend upon morphological properties of the inclusion such as its aspect ratio ($a_r = \frac{\text{length}}{\text{diameter}} \approx 1$ for particles, $\gg$10 for fibres or whiskers and $<$0·1 for platelets) its surface state (smooth, irregular) and on material properties after synthesis such as the chemistry of the inclusion–matrix interface and the stresses generated in the matrix, the inclusion and at the interface[1,2] during sintering.

During fabrication of ceramic composites by uniaxial hot pressing, grain orientation with respect to the pressing direction can occur. As a consequence, the mechanical properties (toughness for instance) can become direction dependent. Indeed, when the reinforcement is obtained by crack deflection or crack bridging mechanisms, the contribution to the toughness will depend on the orientation of the interacting grain with respect to the crack plane. Moreover, the interaction of the crack with the reinforcing particle will also depend on its morphology. For instance, the orientation dependency of the contribution to reinforcement of one-dimensional fibre-like grains can be quite different compared to two-dimensional platelets.

In this paper, we will discuss the anisotropic properties of a hot pressed platelet reinforced silicon nitride composite compared to the monolithic material. In those materials, reinforcement can mainly be attributed to reinforcing mechanisms such as crack deflection and crack bridging by grains with either high or low aspect ratio (silicon nitride or platelets, respectively). The anisotropy of the mechanical properties of the monolithic material and the composite will be assessed by Scanning Electron Microscopy (SEM) and Transmission Electron Microscopy (TEM) observation as well as by X-ray textural analysis. Taking into account those analyses, the variation of mechanical properties will mainly be discussed in terms of geometrical aspects of the toughening such as the grain orientation and the grain morphology (elongated β-silicon nitride grains or disk-like platelets).

2 Experimental

Composite materials were fabricated by hot pressing (HP), using LC12S (H.C. Starck, Germany) commercial silicon nitride powders and 10 vol% of

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silicon carbide platelets (grade SF from C-axis Technologies, Canada). The materials were densified in the presence of sintering aids (1·5 wt% alumina–5·5 wt% yttria) under 20 MPa load and using a 0·1 MPa nitrogen atmosphere.

The microstructure of the densified materials was studied by TEM analysis. Crack interaction both at silicon nitride–silicon nitride and silicon carbide–silicon nitride interfaces was studied by TEM on samples, first, indented and, then, ground and ion thinned at the opposite side.

During hot pressing, anisotropic stresses appear along the compressive z axis (Fig. 1), which can lead to microstructural orientation during densification of the material. The microstructural orientation of silicon nitride grains and platelets was assessed by microscopical observation, textural analysis by X-ray diffraction and by TEM analysis.

The toughness (K_{IC}) of the monolithic material and the composites was assessed by the SENB technique, for different, perpendicular notch orientations with respect to the hot pressing axis. Four data at least were used for the calculation of the mean toughness values. Crack propagation was studied in different directions with respect to the hot pressing axis by indentation experiments.

3 Results and discussion

In order to assess the reinforcing mechanism of the composite, we first studied its microstructure. Fig. 2, showing a TEM micrograph, reveals a platelet orientated in a plane perpendicular to the c axis direction, embedded in a silicon nitride matrix consisting of interlocked elongated β-Si_3N_4 grains. We can observe that the (001) basal platelet plane, in spite of the presence of a high stacking fault density, looks smooth, just the opposite of whisker composites where microfaceting of the whiskers leads to an irregular shaped interface.[3] Therefore, platelets appear to be very suitable for crack deflection—or bridging-mechanisms, compared to whiskers. Kreher *et al.*[4] have studied the microstructural residual stresses in SiC-platelet reinforced silicon nitride. They showed that the Si_3N_4 matrix is slightly under compression, while tensile stresses basically appear at the particle–matrix interface and in SiC platelets which undergo tension ranging from 100 to 300 MPa.

The interfacial tensile stresses are, a priori favourable for crack deflection reinforcement.

TEM observation of the platelet–matrix interface locally showed glassy phase at grain boundaries (Fig. 3) and, probably, the presence of graphite (Fig. 4), which, according to Braue,[3] who studied the same kind of composites by HREM, could be related to excess carbon at the SiC–platelet surface.

The smooth and weakly bonded platelet–matrix interface is favourable for reinforcing mechanisms based on debonding at the interface which was confirmed by TEM analysis of indented composites (Fig. 5). Moreover, debonding also appears to occur in the Si_3N_4 matrix (Fig. 6).

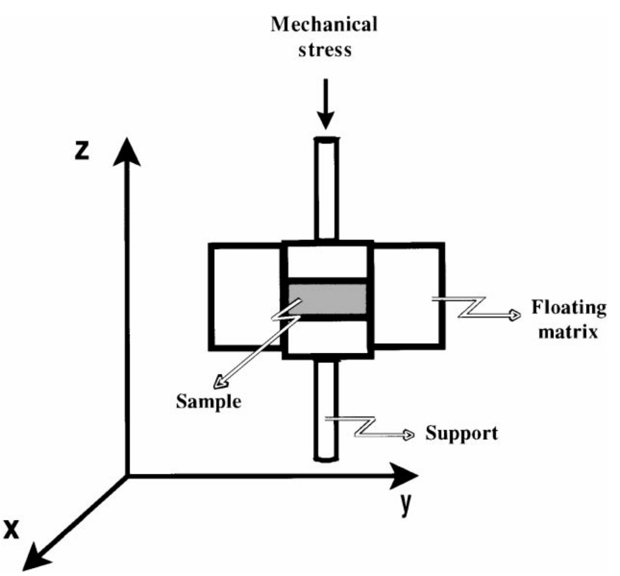

Fig. 1. Experimental hot pressing configuration with respect to a framework.

Fig. 2. TEM micrograph of composite showing a platelet in a plane perpendicular to the c axis.

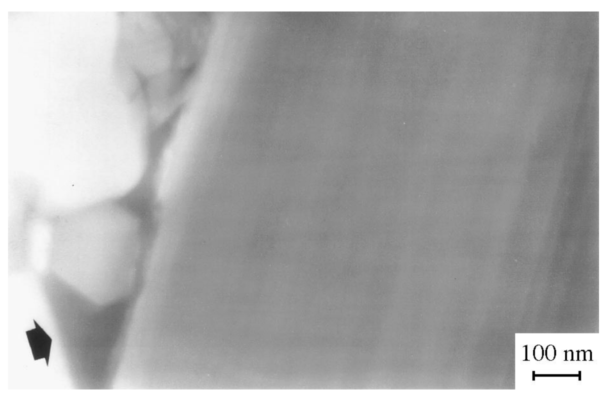

Fig. 3. TEM observation of the platelet–matrix interface showing glassy phase.

Debonding of the platelet–matrix interface or the silicon nitride grain boundary is favourable for crack deflection and bridging mechanisms. The orientation of these grains with respect to the crack tip will have a major impact on the reinforcement degree. Obviously, they should preferentially be orientated with their largest dimension perpendicular to the crack propagation direction. Therefore, the microstructural orientation of both Si_3N_4 and SiC grains with respect to the compressive axis was assessed.

Optical observations, together with X-ray textural analysis both showed that the platelets are orientated with their c axis parallel to the compression z axis, whereas their (001) plane, of 6H polytype, is in a plane perpendicular to this axis. The β-Si_3N_4 grains also show texturing. Indeed, X-ray textural analysis shows the grains to be orientated with their c axis in the compressive (x,y) plane. This was confirmed by TEM microstructural observation (Figs 7 and 8). Moreover, from these micrographs, the aspect ratio of the β-Si_3N_4 grains, as well as the number of grains having their c axis perpendicular to the plane of observation, was determined in a cross-section parallel to the z axis and perpendicular to it (Fig. 9).

The results, summarised in Table 1, are further evidence for the microstructural orientation of the elongated β-Si_3N_4 grains with their c axis perpendicular to the compressive axis.

The mechanical properties were measured following two perpendicular directions. The notch cut for each kind of K_{IC} measurement by the SENB technique is represented in Fig. 10. The toughness results in Table 2 show that both the monolithic material and the composite show anisotropic properties. Moreover, the toughness of the composite is higher than that of the monolithic material.

The anisotropy of the mechanical properties of the silicon nitride material is explained by a preferential orientation of the β-Si_3N_4 grains. The K_{IC} increase in the case of crack growth in a plane

Fig. 4. TEM observation of the platelet–matrix interface probably showing the presence of graphite.

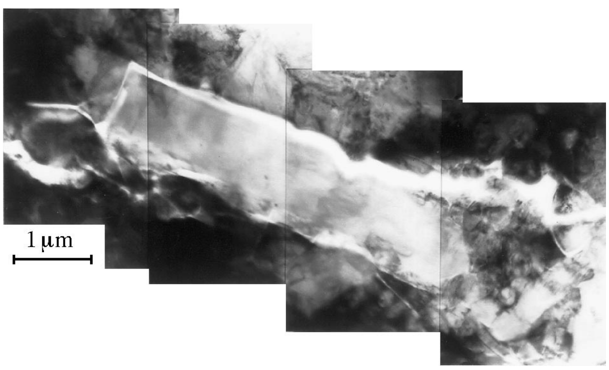

Fig. 5. TEM observation showing crack deflection by SiC platelet.

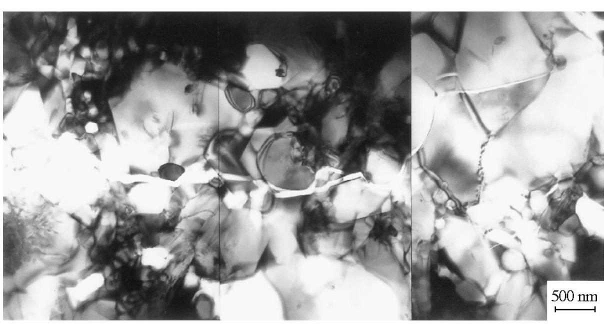

Fig. 6. TEM observation showing crack deflection by Si_3N_4 grains.

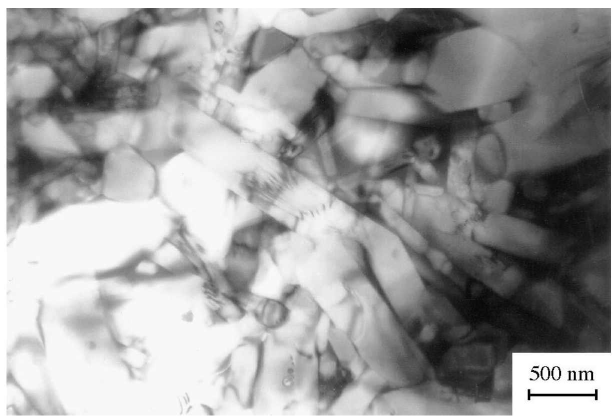

Fig. 7. TEM micrograph of the Si_3N_4 matrix in the (x,y) plane perpendicular to the compressive axis.

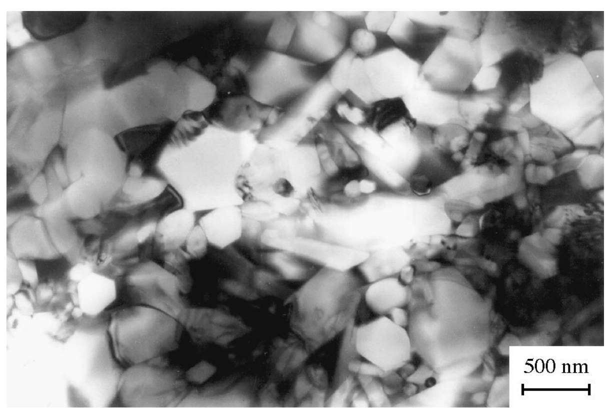

Fig. 8. TEM micrograph of the Si_3N_4 matrix in the (y,z) plane parallel to the compressive axis.

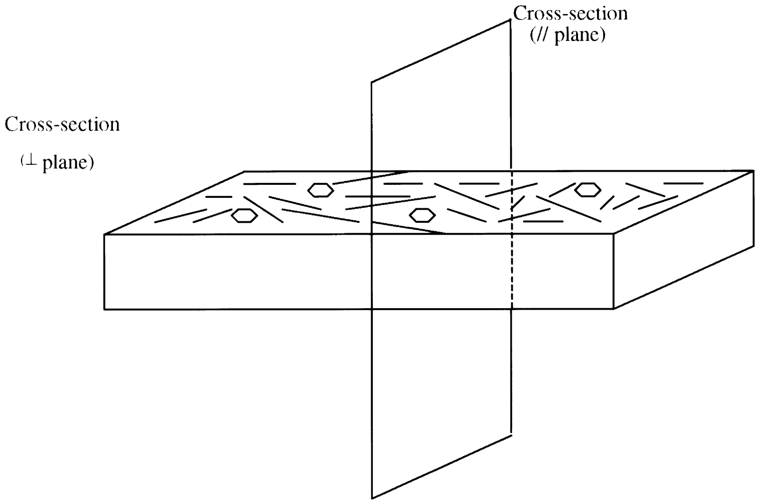

Fig. 9. Definition of cross-sections for microstructural TEM observation.

Table 1. Aspect ratio and number of orientated β-Si_3N_4 in two perpendicular planes (see Fig. 9)

	Aspect ratio	*Number of grains having their c axis ⊥ to the cross-section/ total number of grains*
⊥ plane	3·2	0·3
// plane	1·6	0·8

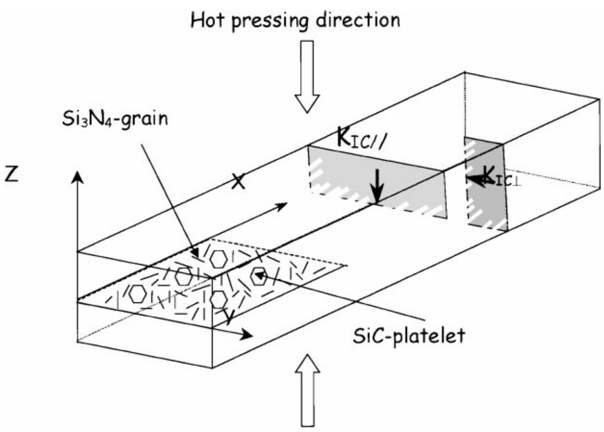

Fig. 10. Representation of notch planes for K_{IC} measurement following different directions by the SENB technique.

Table 2. Critical stress intensity factor of monolithic material and composite measured in a plane parallel to the hot-pressing axis and a direction parallel to the axis (K_{IC} parallel) or perpendicular to the axis (K_{IC} perpendicular)

Material	K_{IC} *parallel (1)*	K_{IC} *perpendicular (2)*	ΔK_{IC} (1)−(2)
LC12s (SN)	6·3 ± 0·3	4·9 ± 0·3	1·4
SN + 10% vol SF	7·7 ± 0·4	6·2 ± 0·2	1·5

parallel to the pressing axis and following a direction parallel to this axis is explained by the fact that the *c* axis of the Si_3N_4 grains are orientated perpendicular to the uniaxial compressive axis and this orientation favours reinforcing mechanisms by crack deflection. Moreover, in the same table, we notice that the difference between critical stress intensity factors (ΔK_{IC}) measured following a parallel or perpendicular direction with respect to the compressive axis can mainly be explained by the anisotropy of the silicon nitride matrix alone ($\Delta K_{IC} \approx$ constant) and is not or little modified by the incorporation of platelets. These observations can be explained by the difference in morphology between platelets and silicon nitride grains. Indeed, the β-Si_3N_4 grains are elongated following one direction, parallel to the *c* axis of the hexagonal network, whereas the α-SiC platelets are elongated following two directions perpendicular to the *c* axis of the hexagonal network. As a consequence, when grains and platelets are preferentially orientated, crack deflection is only possible following one direction in the case of the silicon nitride grains and following two perpendicular directions in the case of the platelets. The reinforcing mechanism is therefore limited to one direction in the case of silicon nitride but is operational following two directions in the case of the platelets. These considerations explain why anisotropic properties are only observed in the case of the silicon nitride matrix, whereas the reinforcement is operational following two directions in the case of the platelets.

4 Conclusions

- Hot pressing of Si_3N_4–SiC platelet composites leads to a material with an anisotropical microstructure consisting of platelets orientated with their basal plane in the compressive plane surrounded by elongated β-Si_3N_4 grains with their *c* axis perpendicular to the compressive axis.

- Due to a weak interface, consisting of glassy phase and graphite at the platelet–matrix grain boundary, platelets and matrix both contribute to the toughening of the composite through crack deviation and, sometimes, bridging mechanisms.
- This reinforcing mechanism highly depends on the grain orientation with respect to the crack plane. Preferentially, the grains should be orientated with their largest dimension perpendicular to the crack plane.
- Due to their orientation, one dimensional Si_3N_4 fibres reinforce only according to one direction whereas two dimensional platelets lead to a higher toughness in two perpendicular directions.

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