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Fracture toughness of Si_3N_4 and its Si_3N_4 whisker composite without sintering aids

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Si_3N_4 without sintering aids is studied with special interest to the fracture behaviour and its relation to microstructure. Cracks propagated almost transgranularly and no rising R-curve behaviour was found, because crack-wake region gave no contribution on toughening due to very high grain-boundary bonding strength. Microstructure with highly elongated grains was obtained by addition of 20% Si_3N_4 whisker, but fracture toughness was found to be similar to that of the monolithic Si_3N_4 with equiaxed grains. It is recognized that fracture toughness is not determined simply by apparent microstructural parameters such as mean aspect ratio of grains when grain-boundary bonding is sufficiently strong. Detailed examination of microfracture behaviour is, therefore, necessary for the analysis of toughening in this kind of composites.

1. Introduction
Fracture toughness as well as strength has been used most frequently as a parameter characterizing engineering ceramics. Silicon nitride and its related nitrogen ceramics often exhibit higher fracture toughness compared with usual engineering oxide-ceramics such as alumina and mullite, and a great deal of work has been done to improve toughness in these systems. Despite this work, tougher materials seem to have been pursued only by empirical means: changing the composition of sintering aids and/or sintering conditions. This may be due to lack of a simple system in which fracture behaviour can be analysed without the influence of complex grain boundary phases.

Recently the present authors and others have succeeded in fabricating fully densified Si_3N_4 from highly pure monolithic powder without sintering aids by hot-isostatic-pressing [1–4], which contained a few per cent of glassy pure silica at grain boundaries [2, 3]. This system can be used as a basic reference material for Si_3N_4-based systems. From this viewpoint, the fracture toughness of the system without additives is examined in the present study.

2. Experimental procedure
The starting powder was 99.99% of purity with respect to cation impurities (E10, Ube) and no sintering aids were added. Its mean particle size was about 0.1 μm. In the composite case, 20 vol % of β-Si_3N_4 whiskers 30 μm in length and 0.5 μm in diameter (SNWB, Ube) were added. Green bodies with about 50% of density were glass (Pyrex, Corning) capsulated and HIPed at 1950 °C and 170 MPa for 2 h. Details of the sintering procedures were described in a previous report [1]. The oxygen content was checked before and after the HIPing and remained unchanged at 1.3 wt%. The microstructure was observed after lapping and etching by NaOH at 380 °C for 1 min. Grain morphology observed in this way after the sintering is shown in Fig. 1. The density of the sintered bodies was higher than 99.8% and only β-Si_3N_4 was detected by X-ray diffraction.

The fracture toughness of the sintered bodies was examined by different techniques using cracks induced by Vickers indentation. Nose and Fujii have recently proposed a use of pop-in precrack for the evaluation of fracture toughness in ceramics from the bending strength and named it the single-edge precracked beam (SEPB) method [5]. In this method, bridge indentation (BI) force is applied until the point of pop-in to the specimen containing controlled surface flaws induced by Vickers indentation. In the present study, specimens of $4 \times 3 \times 18\ mm^3$ were indented by Vickers load of 196 N at the centre of the tensile

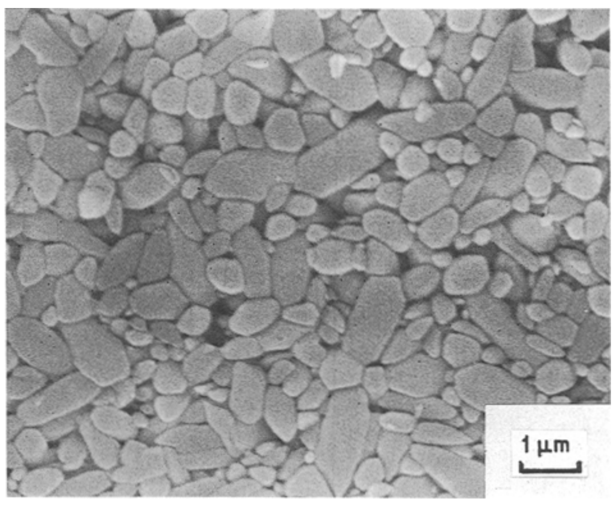

Figure 1 SEM micrograph of lapped and etched surface.

surface. The specimens were then set in a BI jig with a detector of acoustic emission (AE) and loaded with a cross-head speed of 0.1 mm min^{-1} in a test-machine (type 1185, Instron). Fig. 2 shows the number of AE events detected as a function of load. A sudden and large increase of AE events detected at around 1.5 kN shows the occurrence of pop-in. For the measurement of fracture toughness, the BI indentation was quitted immediately after the pop-in and the specimen was subjected to three-point bending by a rate of 0.5 mm min^{-1}. The outer span was 15 mm. After the bending, precracking length, a_p, was measured by optical microscope and the fracture toughness was calculated from the bending strength σ_f by the following equation

$$K_{Ic} = Y\sigma_f a_p \quad (1)$$

Geometrical constant Y was obtained by the equation given by Wakai *et al.* [6] as a function of a_p/w and l/w, where l and w are width of specimen and outer span for a three-point bending test, respectively. Obtained K_{Ic} value was 1.8 ± 0.2 MPa m$^{1/2}$, which may be one of the lowest values observed in fully dense polycrystalline Si_3N_4.

3. Results

In polycrystals, the contribution of microstructure for toughening can be evaluated by the resistance-curve (R curve) behaviour which appears in materials whose apparent fracture resistance increases as crack propagates because of shielding force originated in the wake region of the crack, for example by bridging effects. The R curve behaviour of the present material was examined by using the median crack induced by Vickers indentation [7, 8]. Fig. 3 shows the crack length, c, measured on the surface of specimen after Vickers indentation of various load, P_V. For P_V greater than 9.8 N, the experimental results can be fitted to a single line of slope 1.51 ± 0.03, which shows the residual stress field in median crack configuration. The fracture toughness can be calculated using an

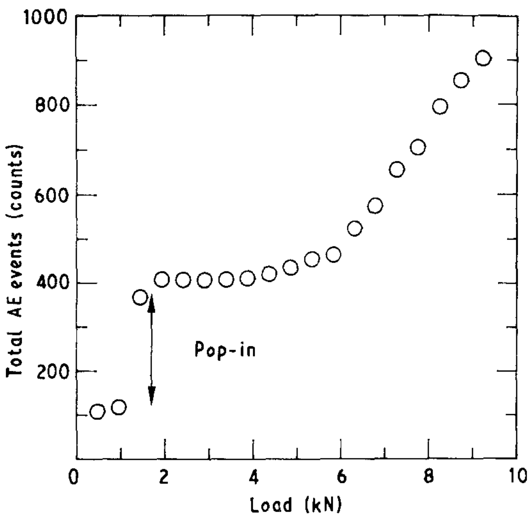

Figure 2 Total acoustic emission events as a function of load during bridge indentation. Vickers indentation of 196 N was made before the test.

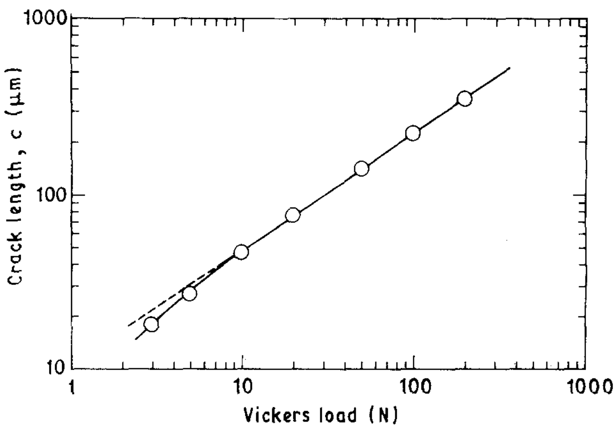

Figure 3 Surface crack length induced by Vickers indentation for 15 s.

empirical equation for median crack [9]. In the present case, the value was 3.0 ± 0.4 MPa m$^{1/2}$.

Fig. 4 shows the bending strength of the present monolithic sintered bodies as a function of Vickers indentation load. The specimens were $3 \times 4 \times 18$ mm^3 in dimension and two or three indentations were made at the centre of the tensile surface. It has been shown [8] from a simple calculation that the bending strength measured as above should be proportional to the $-1/3$ power of the Vickers indentation load in the absence of R curve behaviour. In the present experiments, the obtained slope was -0.36 ± 0.02, indicating independence of the value of fracture resistance on crack extension length. The fracture resistance was calculated to be 2.1 ± 0.2 MPa m$^{1/2}$ by using the following equation

$$K_R = k(\Delta a)^m \quad (2)$$

where Δa is the crack extension length, and constants k and m can be calculated from the slopes of Figs 3 and 4 by assuming a hemispherical crack.

The observation of the crack propagation profile can explain the fracture behaviour of this system. Fig. 5 shows a crack induced by Vickers indentation at 49 N. As can be seen, the crack propagated almost transgranularly. Although it was deflected slightly at some grain boundaries, it cannot propagate intergranularly. As a result, it entered the neighbouring grain. This must be due to very high bonding strength

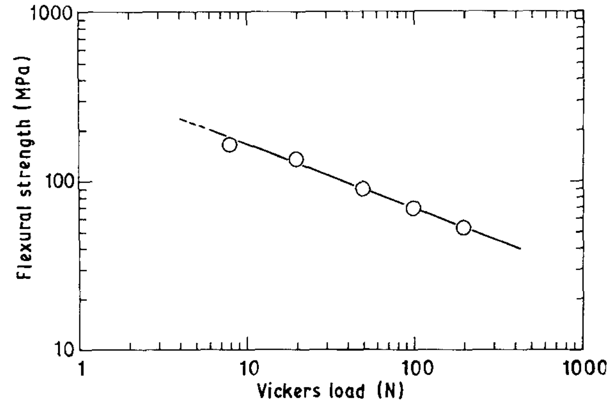

Figure 4 Three-point bending strength for specimens after Vickers indentation.

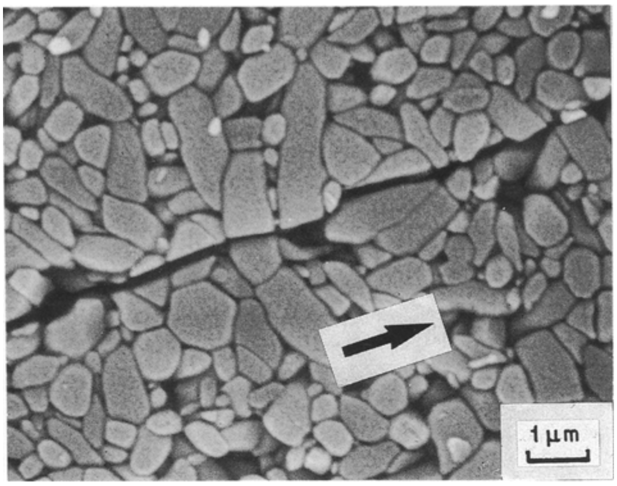

Figure 5 Crack profile induced by Vickers indentation at 49 N on lapped surface. Specimen was etched after the indentation.

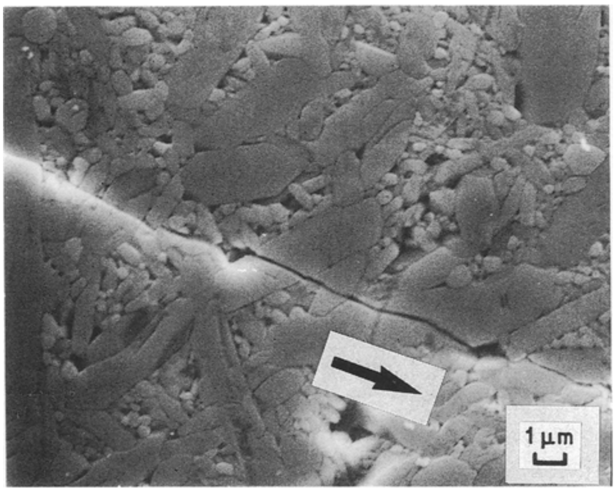

Figure 6 Crack profile in Si_3N_4–20 vol % Si_3N_4 composite sintered without additives.

of grains in this material. In this condition, the wake region is unable to give significant contribution to toughening. Consequently, the toughness of this polycrystal increased not so much with respect to single crystal value and the increment can be ascribed to the interaction between crack tip and microstructure. High resolution electron-microscopy observation revealed that grain boundary phase of this material is a thin layer of pure silica about 1.2 nm in thickness [2]. Such a small thickness may be one of the main reasons for the high bonding strength.

It was shown [10] that fracture toughness of hot-pressed Si_3N_4 is a linear function of mean aspect ratio of grains, and this result was utilized to explain the fracture behaviour of this material from a crack-deflection viewpoint [11]. These analyses were based on the assumption that fracture mode is perfectly intergranular. For materials showing a high degree of transgranular fracture such as the present systems, the high aspect ratio of grains may not be very useful. An experimental evidence can be obtained in a Si_3N_4–20 vol % Si_3N_4 whisker composite sintered without additives under exactly same conditions as the above. Fig. 6 shows the grain morphology of the composite and the crack propagation profile through it. As is clearly seen, the crack cuts the whisker and propagates almost transgranularly. As a result, fracture toughness in the composite is 3.5 ± 0.4 MPa $m^{1/2}$ by the indentation method, which is not very different from the matrix value, 3.0 MPa $m^{1/2}$, despite a microstructure with sensibly higher mean aspect ratio. Detailed analysis of Si_3N_4 without additives reinforced by whiskers will be described elsewhere [12].

4. Conclusion

In summary, the present system demonstrates well the importance of the study of grain boundary properties as well as grain morphology. Examination of mean aspect-ratio of grains or frequency of crack deflection without taking into consideration the grain–boundary bonding strength may lead to insufficient conclusions. Special care should be paid to a comparison of systems sintered with different additives or reinforcements.

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