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Mechanical properties of Si_3N_4 ceramics reinforced with SiC whiskers and SiC platelets

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Silicon nitride ceramics reinforced with SiC whiskers and SiC platelets were fabricated by hot pressing and their mechanical properties were studied. They showed higher fracture energy than conventional composites, particularly when they were consolidated by gas-pressure hot pressing at high temperature. A high fracture toughness (10.7 MPa $m^{1/2}$) which was measured by the single-edge pre-cracked beam method was achieved. Furthermore, a unique method to observe the crack propagation behaviours directly in a scanning electron microscope with loading devices was developed. As a result, much bridging and pull-out of the whiskers and the elongated Si_3N_4 grains, and crack deflection along the platelets, were observed behind the crack tip. This means that these grains are effective in enhancing the fracture resistance during crack propagation. © *1998 Chapman & Hall*

1. Introduction

Si_3N_4 ceramics have good mechanical properties at high temperatures and are, therefore, considered as a candidate material for application to high-temperature gas turbines. However, a catastrophic fracture can occur because the strength is very sensitive to flaws in the materials. Therefore, it is important to improve the fracture toughness and to enhance the reliability. It is well known that the fracture toughness of Si_3N_4 increases on raising the sintering temperature because the Si_3N_4 grain morphology changes from granular to needle like according to the α-to-β transformation [1–3], and it can also be increased by adding second phases, such as whiskers [2, 4–6] or platelets [7, 8]. Regarding whiskers, however, it is difficult to achieve uniform dispersion, especially in a high volume content. On the other hand, platelets give better fracture resistance than whiskers although they reduce the strength.

In this study, we focused on Si_3N_4 composites reinforced with both SiC whiskers and SiC platelets, and the mechanical properties were investigated.

Furthermore, in order to clarify the toughening mechanism, we tried to observe the fracture phenomena directly. A unique method which realized stable crack propagation and direct observation during fracturing using scanning electron microscopy (SEM) is presented.

2. Experimental procedure

2.1. Sample preparation

SiC whiskers (Tokai Carbon Co. Ltd; TWS-400), SiC platelets (C-Axis Technology; SF grade) and Si_3N_4 powders (Ube Ind. Ltd; SN-E10), were used for fabricating the composites. The diameter and length of the whiskers used were about 1 μm and 30–50 μm, respectively, the diameter and thickness of the platelets used were about 10–20 μm and 1 μm, respectively. The content of each reinforcement was varied in the range 0–30 wt % and some samples including both reinforcements were also prepared. 5 wt % Y_2O_3 and 5 wt % $MgAl_2O_4$ powders were used as sintering additives. The powders were mixed by ball milling in butanol for 24 h. The slurries were dried, and the starting powders were cold pressed. The powder compacts were hot pressed at 1650–1770 °C for 1 h at a pressure of 30 MPa in a nitrogen atmosphere. Some of the compacts were hot pressed at 2000 °C for 2 h under nitrogen gas pressure of 8 atm for the purpose of Si_3N_4 grain growth.

Samples were machined to the specimens for the single-edge notched-beam (SENB) and chevron notched-beam (CN) tests. The sizes and configurations of the specimens are shown in Fig. 1a and b. The notch width was 0.1 mm. The single-edge pre-cracked beam (SEPB) specimens were made by introducing pop-in cracks into SENB specimens with a notch 0.2 mm in depth [19].

2.2. Characterization

The densities of the samples were measured by the Archimedes method, and Young's modulus was determined by the acoustic pulse method.

The flexural strength, σ_f, and fracture toughness, K_{Ic}, were measured in air at room temperature using an Instron testing machine (1361 type). Flexural strength measurements were performed using

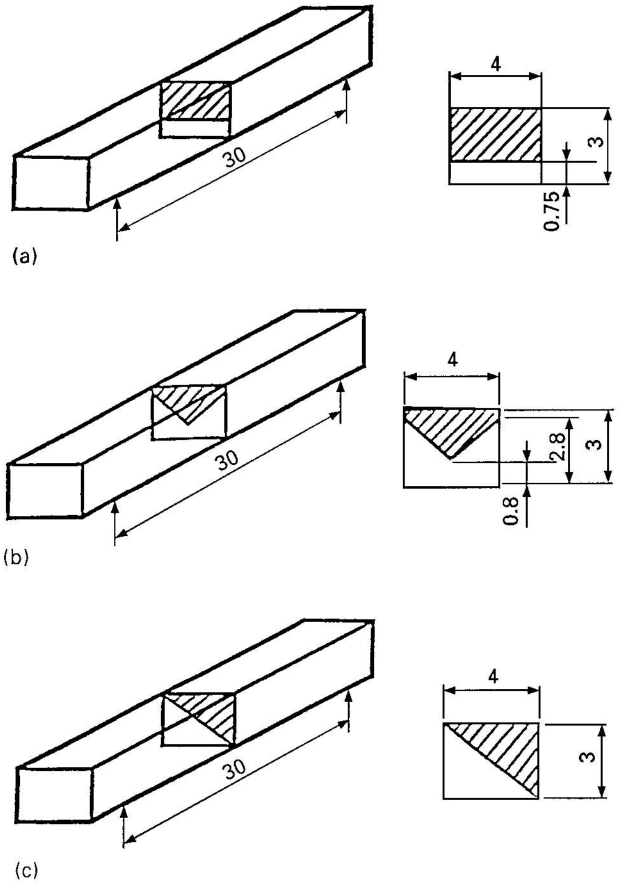

Figure 1 Configurations and dimensions of specimens in millimetres: (a) SENB specimen; (b) CN specimen; (c) modified CN specimen.

three-point loading with 30 mm span. K_{Ic} was measured by the SENB and SEPB methods using four-point loading. The cross-head speed was 0.5 mm min^{-1} for both σ_f and K_{Ic} measurement. Moreover, the effective fracture energy, Γ_{eff}, was evaluated from the work of fracture, which is calculated from the areas under the load–displacement curves, measured by the CN test method at a cross-head speed of 0.05 mm min^{-1}.

The microstructure was investigated by SEM after the Si_3N_4 grains were removed by reactive ion etching.

2.3. Observation of crack propagation behaviour

To investigate the effect of toughening, we attempted to observe directly the crack propagation behaviours. Modified CN specimens with a right-angled triangle cross-section as described in Fig. 1c were used. On increasing the bending load on the specimen, the crack becomes visible on the side of the specimen which is coated with gold after polishing and removal of grain-boundary phase by chemical etching. This specimen was set on the stage in the scanning electron microscope (Fig. 2) and loaded by motor driving at a cross-head speed of 0.05 mm min^{-1}. During the test, SEM images were recorded on video tapes and the applied load was monitored.

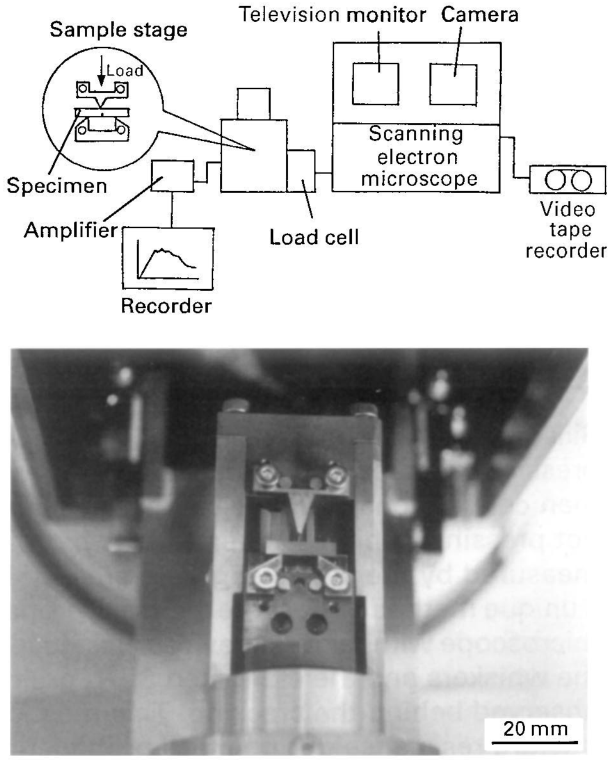

Figure 2 A modified CN specimen was set in the loading equipment on the scanning electron microscope sample stage. The crack propagation behaviour was observed *in situ* by applying a three-point bending load.

3. Results and discussion

3.1. Mechanical properties

Most of the samples reached the full density. The flexural strength of the materials including either whiskers or platelets is shown in Fig. 3 as a function of SiC platelet content or SiC whisker content. The flexural strength of the platelet composites decreased with increasing platelet content; however, that of the whisker composites did not decrease. This probably means that the platelets are more prone to be the origin of the fractures than the whiskers are (Fig. 4). Fig. 5 shows the fracture energy, Γ_{eff}, of the materials reinforced with SiC whiskers or platelets as a function of the content. It was confirmed that the platelets were more effective than the whiskers in improving Γ_{eff}, particularly when the hot-pressing temperature was high.

The mechanical properties of the composites reinforced with both whiskers and platelets are shown in Fig. 6. In this figure, w5, w10, w20 and w30 indicate the data from the samples with 5, 10, 20 and 30 wt % SiC whiskers, respectively. The flexural strength decreased with increasing SiC platelet content and did not depend on the whiskers in the composite. The fracture toughness measured by the SENB method also decreased slightly with increasing platelet content. However, the composite with both whiskers and platelets which is hot pressed at 2000 °C showed improved toughness. The fracture toughness values measured by the SEPB method are listed in Table I, which shows different results from those obtained by the SENB method. The toughness values measured by SEPB of the composites hot pressed at 2000 °C were

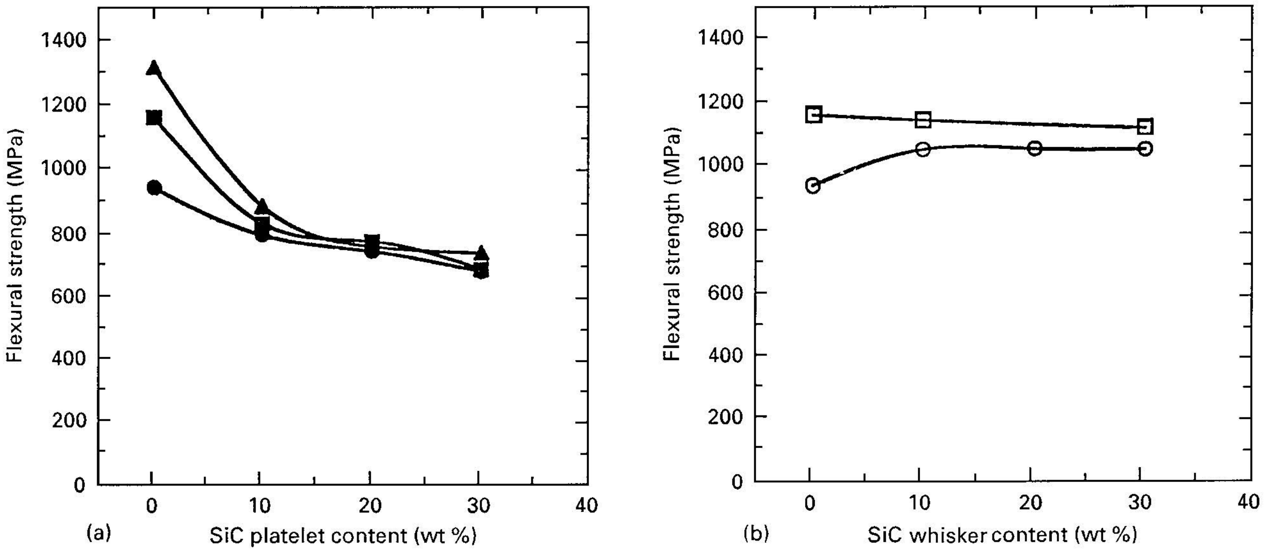

Figure 3 The flexural strength of Si_3N_4 composites at room temperature for various hot-pressing temperatures as a function of (a) SiC platelet content ((■), 1770 °C; (▲), 1700 °C; (●), 1650 °C) and (b) whisker content ((○), 1650 °C; (□), 1770 °C).

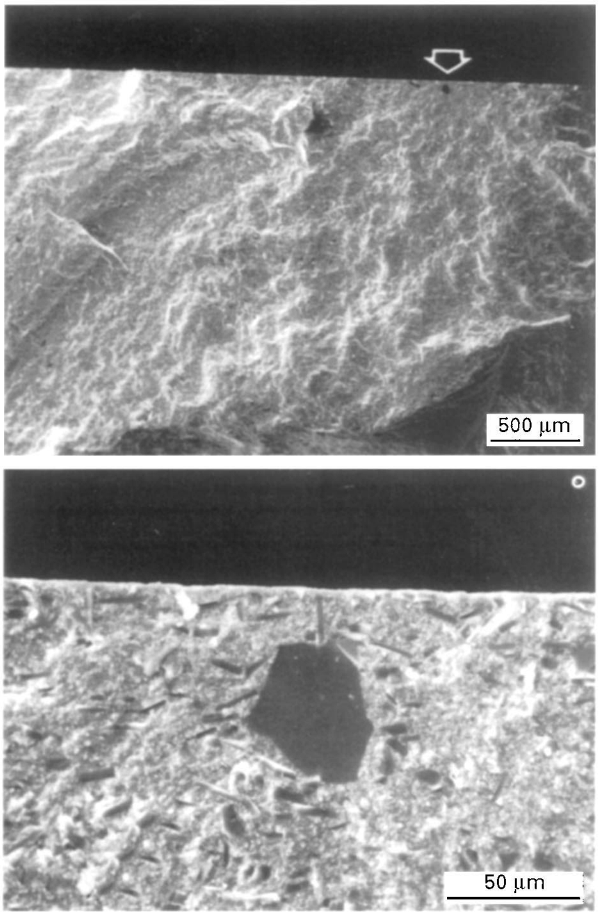

Figure 4 Scanning electron photographs of platelet composite. Platelets are more prone to be the origin of fractures.

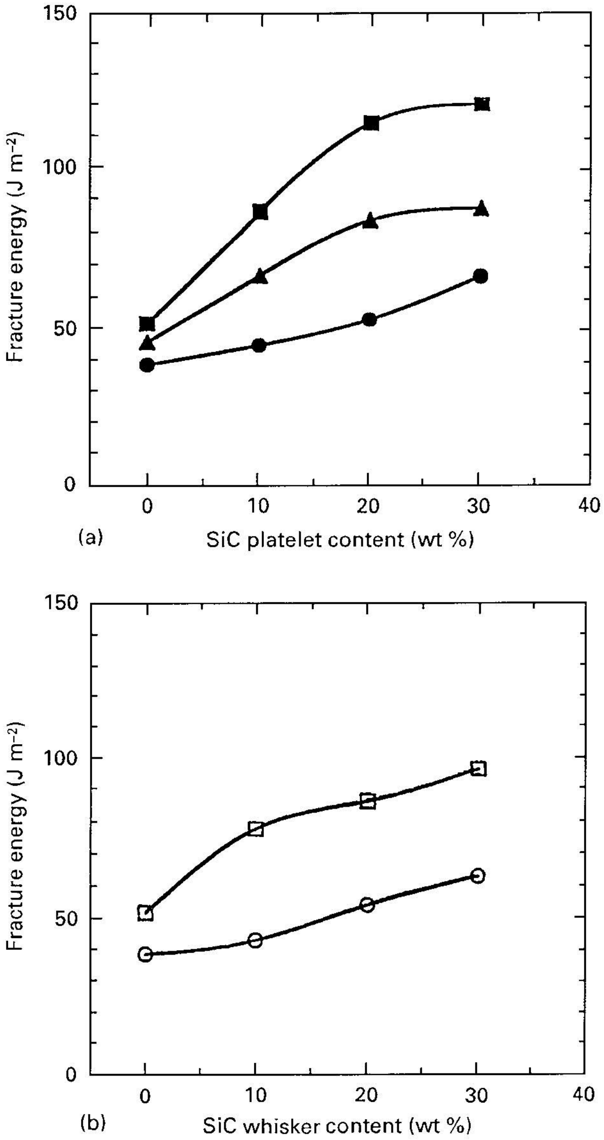

Figure 5 Effects of SiC whisker or SiC platelet content on the fracture energy, Γ_{eff}, at room temperature for various hot-pressing temperatures as a function of (a) SiC platelet content ((■), 1770 °C; (▲), 1700 °C; (●), 1650 °C) and (b) SiC whisker content ((○), 1650 °C; (□), 1770 °C).

higher than those at 1770 °C, and the composite with 10 wt % whiskers and 20 wt % platelets showed the highest value of 10.7 MPa m$^{1/2}$. Considering the features of the SEPB method, the pop-in pre-crack may include the grain interlocking zone behind the crack tip before the fracture test. In fact, it is thought that this interlocking effect caused the high K_{Ic} value for the SEPB method. Fig. 7 shows the scanning electron micrographs of monolithic Si_3N_4 and composite

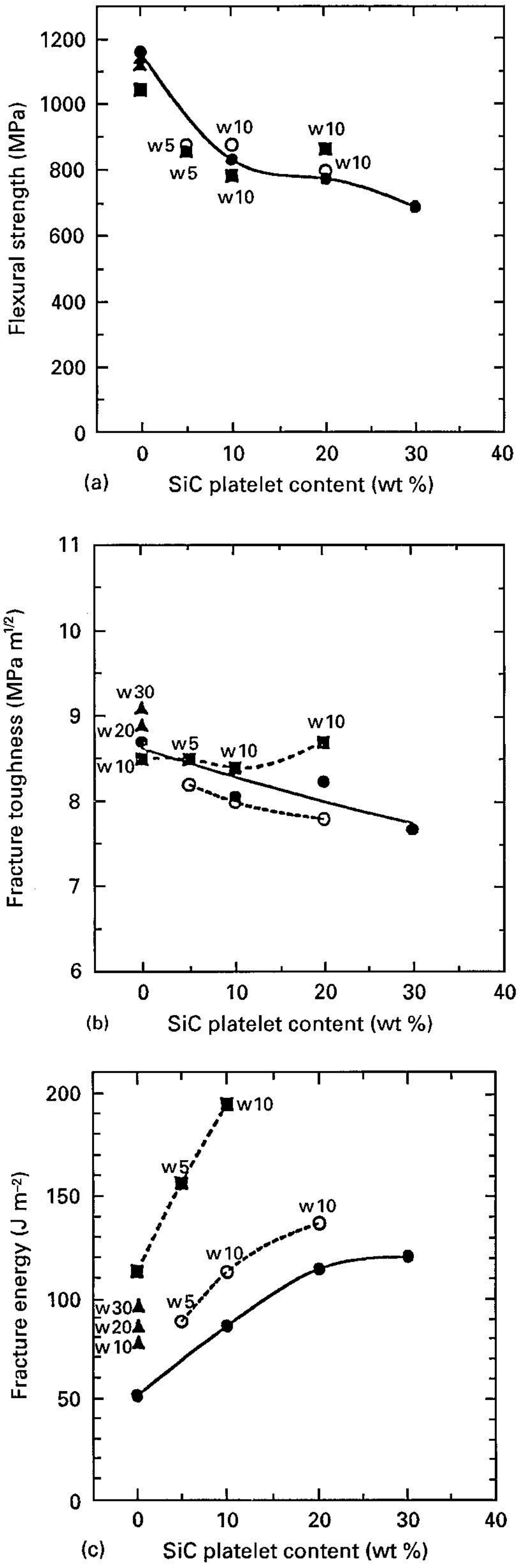

Figure 6 (a) Flexural strength, (b) fracture toughness and (c) fracture energy as functions of platelet content. (●), 1770 °C platelets; (▲), 1770 °C whiskers; (○), 1770 °C platelets + whiskers; (■), 2000 °C platelets + whiskers; (——), data for materials reinforced with only SiC platelets.

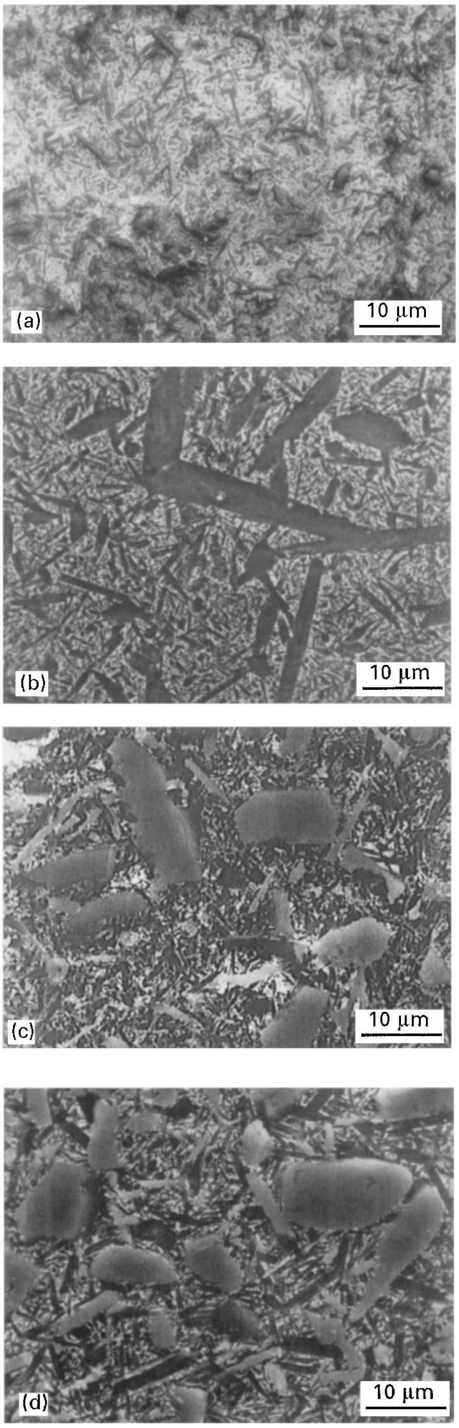

Figure 7 Scanning electron micrographs of (a), (b) monolithic Si_3N_4 and (c), (d) Si_3N_4 composite reinforced with both 10 wt % SiC whiskers and 20 wt % SiC platelets: (a), (c) material hot pressed at 1770 °C; (b), (d) material hot pressed at 2000 °C under gas pressure. The Si_3N_4 grains were etched before the grain-boundary phase and SiC grains. The non-etched grains are estimated to be SiC whiskers or SiC platelets.

reinforced by 20 wt % platelets and 10 wt % whiskers. The Si_3N_4 grains were etched before the grain-boundary phase and platelets. The microstructure of the monolithic Si_3N_4 depends on the hot-pressing temperature and elongated Si_3N_4 grains are observed in the material fabricated by gas-pressure hot pressing at 2000 °C. On the other hand, the grain growth of Si_3N_4 seems to be restrained for the whisker and platelet composites (Fig. 7c and d). In spite of the different microstructures of the materials hot pressed at 2000 °C, their fracture toughnesses measured by the SEPB method were almost the same and high (about 10 MPa $m^{1/2}$). It is thought that the high toughness of the monolithic material was mainly owing to the elongated Si_3N_4 grains and that of the composite was mainly owing to whiskers and platelets. Furthermore,

TABLE I Fracture toughness K_{Ic} evaluated by the SEPB method

Hot-pressing temperature (°C)	Whisker content (%)	Platelet content (%)	K_{Ic} (SEPB) (MPa m$^{1/2}$)
1770	0	0	6.5
1770	5	5	7.9
1770	10	10	8.5
1770	10	20	9.1
2000	0	0	10.2
2000	5	5	—
2000	10	10	9.0
2000	10	20	10.7

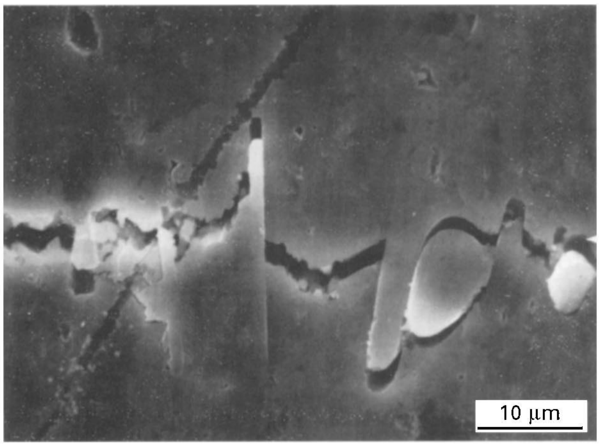

Figure 8 Scanning electron micrograph showing the crack propagation behaviour obtained by *in-situ* observation. Pull-out due to the fibrous grains and crack deflection due to the platelets are seen.

Γ_{eff} for the composites is higher than Γ_{eff} for monolithic Si_3N_4 (Fig. 6c), and, in particular, it is remarkable when hot pressed at 2000 °C. This result shows the effect of the whiskers and platelets on the fracture energy of the composites hot pressed at 1770 °C and the synergistic effect of the elongated Si_3N_4 grains, SiC whiskers and SiC platelets on the fracture energy of the materials hot pressed at 2000 °C. Γ_{eff} shows the fracture resistance during crack propagation. Therefore, it is suggested that these grains were more effective in improving the crack propagating resistance than the crack initiation resistance. Thus, further grain growth of Si_3N_4 for the whisker–platelet composites is expected to result in a higher fracture toughness and fracture energy.

3.2. Observation *in situ*

According to the observation *in situ*, only the whisker-and-platelet-reinforced composite fabricated by gas-pressure hot pressing at 2000 °C did not fail catastrophically. This means that it has a very high fracture resistance. The first crack stopped before the maximum load and propagated again after a short time when the load began to decrease. However, it did not fail completely. The loading device in the scanning electron microscope and the design of the specimen (Fig. 1c) were found to be very useful to observe the crack propagation for materials with a relatively high fracture resistance. Fig. 8 is a typical scanning electron micrograph showing the regions behind the crack tip. Pull-outs of the whiskers and the elongated Si_3N_4 grains and many crack deflections along the platelets could be observed. It is thought that the synergistic effect caused by these grains behind the crack tip, which restrain the crack extension, brought about the high K_{Ic} (SEPB) and the high fracture energy.

4. Conclusions

We reached the following conclusions from the results of this investigation.

1. Materials reinforced by both whiskers and platelets, which are hot pressed at 2000 °C, showed the highest fracture toughness, 10.7 MPa m$^{1/2}$. In particular, they showed a much higher fracture energy than the conventional composites.
2. Observation *in situ* of crack propagation behaviour revealed that the high fracture toughness and energy were caused by bridging or pull-out of the whisker and the elongated Si_3N_4 grains, and by crack deflection along the platelet behind the crack tip. This suggests the existence of high fracture resistance during crack propagation.
3. The loading device in the scanning electron microscope and the specimen design were found to be very useful to study crack propagation behaviours.

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