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Mechanical properties and microstructure of a Si_3N_4/Ti_3SiC_2 multilayer composite

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Abstract

A new Si_3N_4/Ti_3SiC_2 laminated composite which was manufactured by the powder metallurgy method is presented. The work of fracture was measured in four-point bending at a crosshead speed of 0.05 mm/min. The results show that the mechanical properties of multilayered composites consisting of Ti_3SiC_2 layers in Si_3N_4 were significantly enhanced. Three-step toughening behaviors were found in the material. The crack deflection is regarded as a dominant toughening mechanism, meanwhile the unique microstructure and properties of Ti_3SiC_2 contributed to the improvement, thus preventing catastrophic failure. © 2002 Elsevier Science Ltd and Techna S.r.l. All rights reserved.

Keywords: B. Microstructure; C. Mechanical properties; Si_3N_4/Ti_3SiC_2; Multilayer composites

1. Introduction

Silicon nitride is considered a potentially useful structural ceramic for its combination of toughness, strength, hardness, and chemical and thermal durability. Unfortunately, the lack of its reliability in mechanical properties has been a critical problem preventing widespread structural applications. Cook et al. [1] suggested that this problem can be overcome by introducing weak interfaces to deflect a growing crack. Following Cook's idea Si_3N_4 ceramic composites with a layered structure have been evaluated [2–7]. These layered composites were reported to present several improved properties (e.g. increase in fracture toughness, crack arrest, multiple cracking, and so forth). To overcome the lack of reliability, the features of layered composites are especially attractive.

In this paper we have successfully fabricated Si_3N_4/Ti_3SiC_2 multilayer ceramic by powder metallurgy and hot-pressing. It is well known that Ti_3SiC_2 is a novel structural/functional material. Hexagonal Ti_3SiC_2 has a layer structure on the atomic scale. Recent studies show that Ti_3SiC_2 combines the merits of both ceramics and metals, such as being electrically and thermally conductive, as well as easy to machine and resistant to thermal shock, high strength, high toughness, high melting points, low density and thermal stability [8–15]. Thus Ti_3SiC_2 is considered to be effective to overcome the lack of reliability. In present research we selected Ti_3SiC_2 as the weak interface to improve the Si_3N_4 toughness without sacrifice the silicon nitride ceramic properties. The microstructure and mechanical properties of the Si_3N_4/Ti_3SiC_2 multilayer ceramic have been evaluated.

2. Experimental procedure

2.1. Raw materials and fabrication processing

Si_3N_4 powder (E-10, UBE Industries, Tokyo, Japan) with MgO (8 mass%, from Shin-etsu Chemical Co., Tokyo, Japan, 99.99% pure) and CeO_2 (2 mass%, from Shin-etsu Chemical Co., Tokyo, Japan, 99.99% pure) were ball-milled in ethanol for 48h. The slurry was stirred-drying with rotary evaporator at 60 °C, the mixture was reground in a mortar and pestle and screened through a 100-mesh sieve. Ti_3SiC_2 powder was synthesized directly from elemental titanium, silicon, and graphite powders with solid-liquid method at 1270 °C. Pure Ti_3SiC_2 powders were obtained [16] by first removing $TiSi_2$ using HF solution, and then heating the powders in air at 500 °C for 5 h, finally washing the powders with hot

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$(NH_4)_2SO_4 + H_2SO_4$ solution. The resulting powder had average particle size 1.0 μm (purity higher 98%). The dried Ti_3SiC_2 and Si_3N_4 starting mixtures were carefully weighed and alternately layered in a steel die (Φ50 mm inner diameter) in a sequence of Si_3N_4/ Ti_3SiC_2/ Si_3N_4/ Ti_3SiC_2/···/Si_3N_4, then were compressed at 100 MPa. The layer thickness of Si_3N_4 and Ti_3SiC_2 were 0.30 and 0.15 mm, respectively. The green compact was hot-pressed in graphite dies at 25 MPa under flowing argon at 1600 °C for 2 h. The heating and cooling rates were 10 and 15 °C/min, respectively.

2.2. Characterization

Specimens for flexural tests were cut and ground from the billets to nominal dimensions of 4×3×36 mm. Flexure strength was measured in a three-point bending test with spans of 30 mm at a crosshead speed of 0.5 mm/min. The prospective tensile surfaces were ground and polished to achieve a mirrorlike surface finish prior to mechanical testing. The work of fracture was measured in four-point bending test with outer and inner spans of 40 and 20 mm, respectively. The bending tests

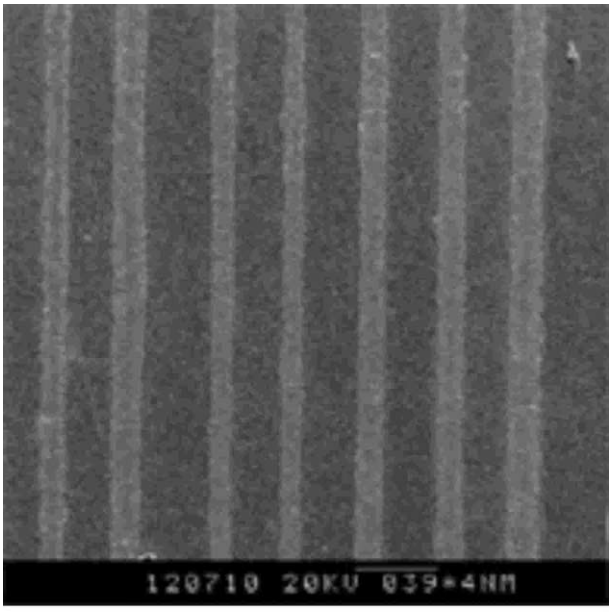


Fig. 1. Macrograph of the laminated composite Si_3N_4/Ti_3SiC_2.

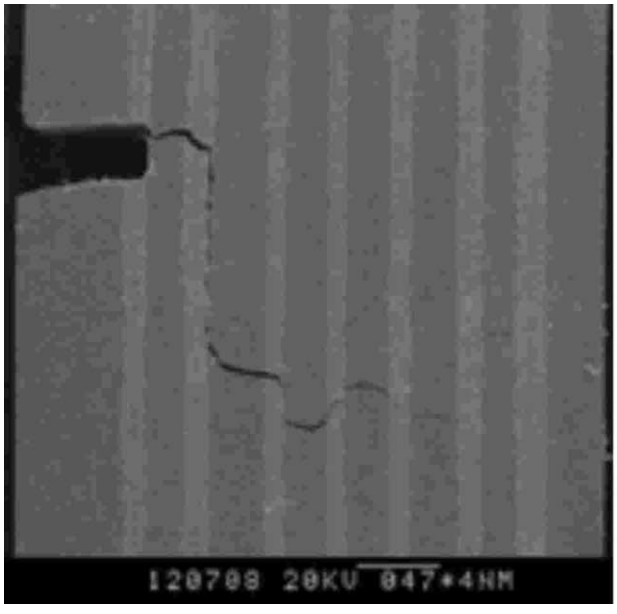
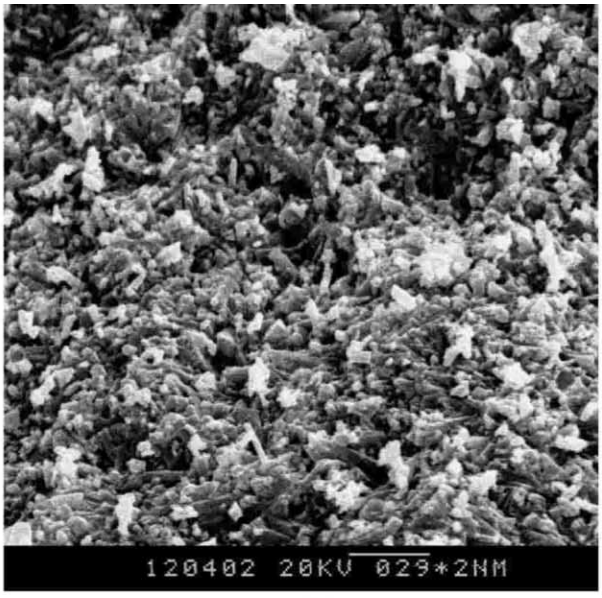
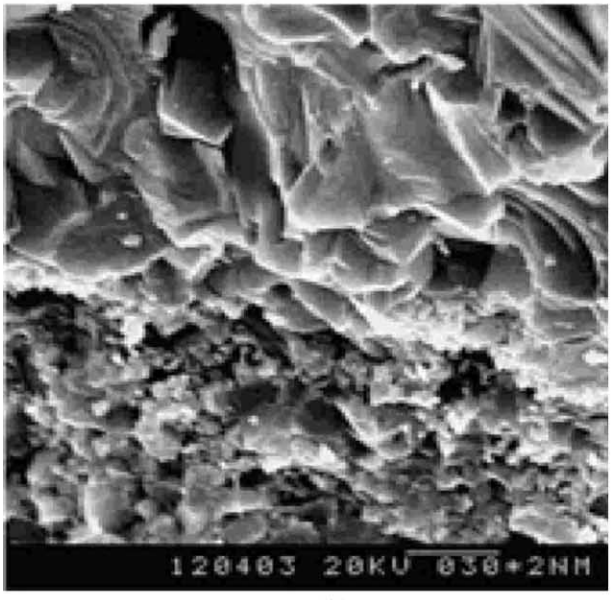


Fig. 2. Crack propagation in the laminated composite Si_3N_4/Ti_3SiC_2.



(a)



(b)

Fig. 3. SEM of the fracture surface of Si_3N_4/Ti_3SiC_2 laminated composites: (a) Si_3N_4 layer; (b) Ti_3SiC_2 layer.

were performed at a crosshead speed of 0.05 mm/min. The work of fracture of each specimen was calculated by determining the area under the load-crosshead deflection curve and dividing it by twice the cross-sectional area of the sample. The fracture surface of the laminated composites were examined by scanning electron microscope (SEM). Energy-dispersive X-ray analysis was used to qualitatively display the compositional distribution in the laminated composites.

3. Results and discussion

The mechanical properties were measured using the hot-pressed laminated sample. The work of fracture of Si_3N_4/Ti_3SiC_2 laminated composites was 4760 J/m^2, and the flexure strength was 746.3 ± 10.2 MPa, while work of the fracture of Si_3N_4 ceramic was only 100 J/m^2. The work of the fracture of Si_3N_4/Ti_3SiC_2 laminated composites was higher than that of Si_3N_4 monolithic materials. Accordingly Ti_3SiC_2 increased reliability of Si_3N_4 ceramic in mechanical properties when it was interface layer, thus preventing catastrophic failure.

On a four-point bending test fixture, conventional ceramics fail by tensile cracking. In contrast, these multilayer ceramics with low interface shear strength could fail by a combination of shear and tensile cracking. The maximum tensile stress in a four-point-bend bar can be expressed as [17]

$$\sigma_{t,\max} = \frac{3ap}{bh^2} \tag{1}$$

where p is the load, a is the distance between the inner and outer loading points, b is width, and h is the thickness of the bar. The relationship between the maximum shear stress and the maximum tensile stress for the four-point load condition can be written as [17]

$$\frac{\tau_{\max}}{\sigma_{t,\max}} = \frac{h}{4a} \tag{2}$$

Shear fracture will occur in these specimens if the maximum shear stress exceeds the shear strength of the interface layer. Based on these considerations, the fracture behavior of a multiplayer ceramic in a bending test depends not only on the microstructure but also on the specimen geometry. The shear cracking would make the specimen more compliant in bending, therefore relieving some of the tensile stresses.

Fig. 1 shows their structure of Si_3N_4/Ti_3SiC_2 laminated composites. The silicon nitride layers appear gray and the Ti_3SiC_2 interfaces appear as thin, bright layers separating the Si_3N_4 layers. Fig. 2 shows a typical crack propagation manner of the hot-pressing laminated composite. As shown in Fig. 2, a major tensile crack propagates through the specimen although it is deflected by Ti_3SiC_2 layers. One remarkable feature observed in Fig. 2 is the interlocking of toothlike, debonded layers. This indicates that the maximum shear stress must exceed the shear strength of the Ti_3SiC_2 layer.

Fig. 3 shows the micrographs of Si_3N_4/Ti_3SiC_2 laminated composites specimen fractured at room temperature. Whisker like β-Si_3N_4 is clearly seen in the Si_3N_4 layer, which indicates α-Si_3N_4 has transformed to the β-Si_3N_4 phase, but sintered temperature was rather

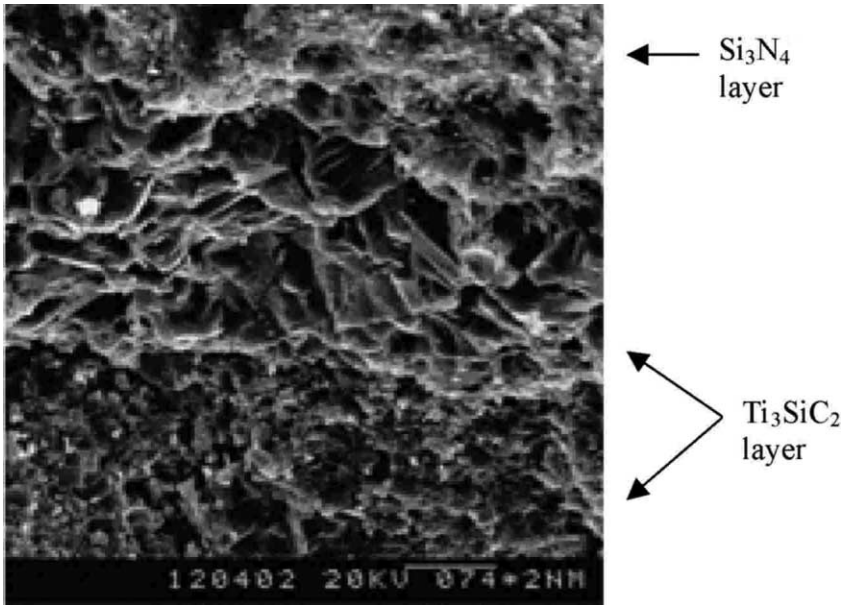


Fig. 4. SEM of Ti_3SiC_2 grains in different positions.

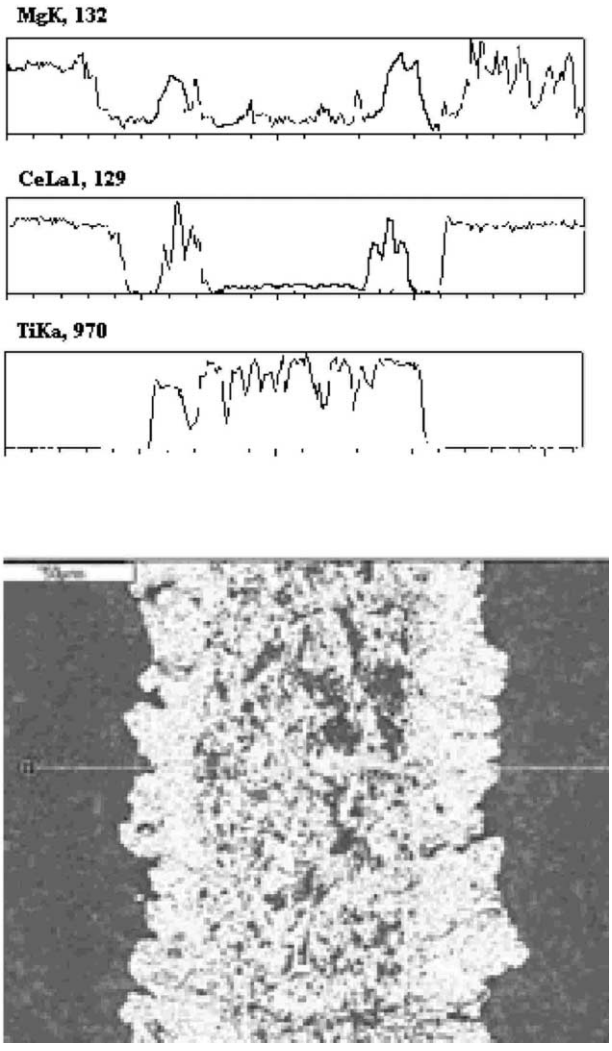


Fig. 5. Element distribution in the laminated composites material.

lowly, so size of the β-Si_3N_4 grains was small. The characteristic of the laminated Ti_3SiC_2 grains can easily be identified in these micrographs. Upon analysis of the fracture surface, the failure in the Ti_3SiC_2 layers appears to be intergranular in nature while exhibiting a highly layered structure. This feature along with the rough nature of the fracture surface has led to the suggestion that this ceramic possesses a weak interface that leads to a behavior of a nanolaminate [18,19]. It is seen that pull-out of Ti_3SiC_2 grains is evidence. Micro-plastic deformation can be seen in such region, which may act more effectively as an energy-absorbing mechanism and toughen the material further.

In the microstructure of laminated composites we found that the size of the Ti_3SiC_2 grains near the Si_3N_4 layer was bigger than for those removed from the Si_3N_4 layer (see Fig. 4). Qualitative energy-dispersive X-ray microanalysis was used to detect the element distribution in the laminated composites material (see Fig. 5). Fig. 5 shows the Mg, Ce and Ti element line map of the region of the laminated material. We found that the Ce and Mg elements have diffused in fringe of Ti_3SiC_2 layers. This indicates that during the sintered process produced liquid in Si_3N_4 layers diffused to Ti_3SiC_2 layers and accelerated growth of the size of Ti_3SiC_2 grains.

4. Conclusions

New Si_3N_4/Ti_3SiC_2 laminated composites have been manufactured by powder metallurgy method. Multilayer materials have shown noncatastrophic failure in four-point bend tests. The three-step toughening behavior observed is a fundamental appearance in Si_3N_4/Ti_3SiC_2 laminated composites. Crack deflection along the weak interfaces was thought to be the major mechanism for improving fracture resistance. The secondary toughening behavior seems to be the layer structure of Ti_3SiC_2 on the atomic scale, pull-out and micro-plastic deformation of Ti_3SiC_2 grains. The difference size of Ti_3SiC_2 grains was tertiary toughening behavior. Multiple toughening mechanisms obviously enhanced fracture resistance. The average apparent work of fracture of the laminated composites was about 4760 J/m^2.

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