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Design, fabrication and properties of layered SiC/TiC ceramic with graded thermal residual stress

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Abstract

The finite element method (FEM) was used to design a symmetrical layered SiC/TiC ceramic with gradual thermal residual stress distribution. In the final model ceramic, the sequence of layers, from surface to inside, was SiC, SiC + 2 wt.% TiC (S2T), SiC + 4 wt.% TiC (S4T), SiC + 6 wt.% TiC (S6T), SiC + 8 wt.% TiC (S8T), and SiC + 10 wt.% TiC (S10T); the thickness ratio of SiC:S2T:S4T:S6T:S8T:S10T was 1:1:1:1:1:10. After the model ceramic had been cooled from assumed sintering temperature 1850–20 °C in FEM calculation, gradual thermal residual stress, varying from surface compressive stress to inner tensile stress, was introduced. The designed ceramic then was fabricated by aqueous tape casting, stacking and hot-press sintering at 1850 °C, under 35 MPa pressure, for 30 min. The surface stress conditions of the sintered ceramic were tested by X-ray stress analysis, and those results were very close to the results from the FEM calculations. Compared with pure SiC and S10T ceramics fabricated by the same process, the designed ceramic showed excellent mechanical properties. The tested strength was close to the theoretical value. The strengthening and toughening mechanisms of the ceramic were ascribed to surface compressive residual stress.

Keywords: Layered ceramic; Mechanical properties; SiC; Tape casting; TiC; FEM

1. Introduction

In the past decade, layered ceramic composites have been studied extensively.[1–5] Ceramic resistance to flaws can be increased by the introduction of compressive residual stress on the surface of layered ceramics, such as those of the ZrO_2/Al_2O_3 system,[6,7] or by crack propagation along the weak interface of laminated ceramics, such as those of the SiC/C and Si_3N_4/BN systems.[8–10]

Surface compressive stress can increase the fracture strength, damage resistance, and fatigue properties, as well as the fracture toughness of layered ceramics.[2,5,6,11] These compressive stresses result mainly from a mismatch of thermal expansion coefficients among layers. In a symmetrical layered ceramic, the design of the residual stress is very important, because the inner tensile stress should be as low as possible when a surface compressive stress is introduced.

In many sandwich structures (A/B/A) or alternate-layered structures (A/B/A/B), residual stress can be calculated by a simple elastic theory.[11] However, when a pure bending load is applied to a structure with a rectangular cross section, the external stress varies linearly from maximum tension on the surface to zero on the central plane.[12] Thus, we believe that thermal residual stress in a symmetrical layered system should be designed to vary gradually from surface compressive stress to central (inner) tensile stress. Therefore, such a structure should have a gradual thermal expansion coefficient change from the surface to the center. In such a case, the finite element method (FEM) is appropriate for designing the structure, because the FEM is a powerful tool for calculating residual stress distribution in composites.[13,14]

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In the present paper, the FEM was used to calculate the thermal residual stress distribution required for optimizing the structure of a layered SiC/TiC composite. The optimized ceramic then was fabricated, and its mechanical properties were tested for comparison with the FEM results.

2. Design of the SiC/TiC composite

A SiC/TiC composite was selected for the present work, because little study has been done on this type of layered material. The physical and elastic properties of SiC and TiC are listed in Table 1. The properties of SiC/TiC composites can be calculated according to the rule of mixtures.[15]

A 2-dimensional initial model for FEM calculation is shown in Fig. 1. The surface layer of the model is SiC, and the inner layer is TiC; the TiC layer is twice as thick as the SiC layer. In view of the symmetry, one-quarter of the model was used for calculation. The right and upper surfaces are free surfaces. All corners were pinned to prevent rigid body translations. The model was assumed to cool from 1850 to 20 °C, with a uniform temperature field. A generalized plane-stress condition was assumed, in which the strain along Z direction was assumed constant. FEM software (MARC 7.1 MARC Analysis Research Corp., Palo Alto, CA) was used for calculation.

Fig. 2 shows the thermal residual stress in the x direction, the σ_{xx} distribution, for one-quarter of Fig. 1. After the model ceramic has been cooled from assumed sintering temperature, a large compressive residual stress was introduced to the surface SiC layer, but the tensile stress in the inner TiC layer was 1350 MPa, far higher than the fracture strength of the TiC material. To decrease the inner tensile stress, the thickness of the inner layer had to be increased or the coefficient of thermal expansion (CTE) mismatch between the surface and the inner layer decreased. To form a gradually varying stress from the surface to the inner layer, SiC + TiC layers of different compositions were inserted between the selected surface and the inner layer. The sintered properties of the SiC/TiC composite also had to be considered. Jiang et al.[16] have reported that the mechanical properties of a SiC/TiC composite decrease when the TiC content exceeds 20 wt.%.

Table 1
Physical and mechanical properties of SiC and TiC

	Elastic modulus (GPa)	Coefficient of thermal expansion (10^{-6}/K)	Density (g/cm^3)	Poisson's ratio
SiC	430	4.8	3.17	0.14
TiC	430	7.4	4.94	0.19

After several steps in view of these above considerations, a final, symmetrical SiC/S10T structure was chosen for the present study, as shown in Fig. 3. The structure modeled in Fig. 3 has the proper surface compressive stress and inner tensile stress. The σ_{xx} distribution of one-quarter of the model is shown in Fig. 4. On the central line of the structure (from points A to B in Fig. 4), the stress gradually varies from compressive to tensile, as shown in Fig. 5.

3. Fabrication of layered SiC/S10T composite and experimental procedure

Various amounts of SiC powder (0.6 μm nominal size, Norton AS Corp., Arendel, Norway) and TiC powder (3 μm nominal size, Zhuzhou Hard Alloy Factory, Zhuzhou, China) were mixed, in deionized water, with 4.8 wt.% alumina (0.48 μm nominal size, Shanghai Wusong Chemical Factory, Shanghai, China) and 3.6 wt.% yttria (4 μm nominal size, Shanghai Yuelong Chemical Factory, Shanghai, China) used as sintering aids. The total volume fraction of the ceramic powders in the slurry was ~50%. A 10% solution of tetramethylammonium hydroxide was used as the dispersant. The mixture was ball-milled for 24 h. Solutions of 10% polyvinyl alcohol (PVA) and glycerol were then added to the slurry as binder and plasticizer, respectively. After the slurry had been ball-milled again for 24 h, it was degassed, using a vacuum pump, then tape cast on a fixed glass plate with a moving blade, at a constant speed of 5 mm/s.

After natural drying, the green sheet was ~220 μm thick. The dried green sheet was cut into rectangles measuring 40×50 mm and stacked according to the sequence in Fig. 3. Four sheets each of SiC, S2T, S4T, S6T and S8T were used (two per side), along with 20 S10T sheets, so that the total number of sheets was 40. The stacked sheets were put into a graphite die,

Fig. 1. The initial SiC/TiC model composite for FEM calculation.

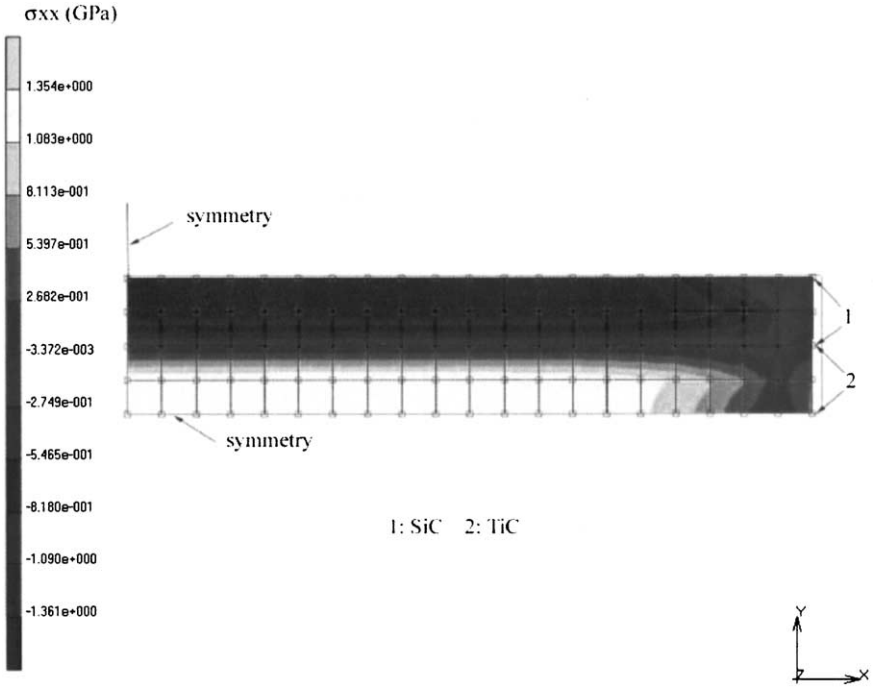

Fig. 2. The σ_{xx} distribution of one-quarter of the initial SiC/TiC model in Fig. 1.

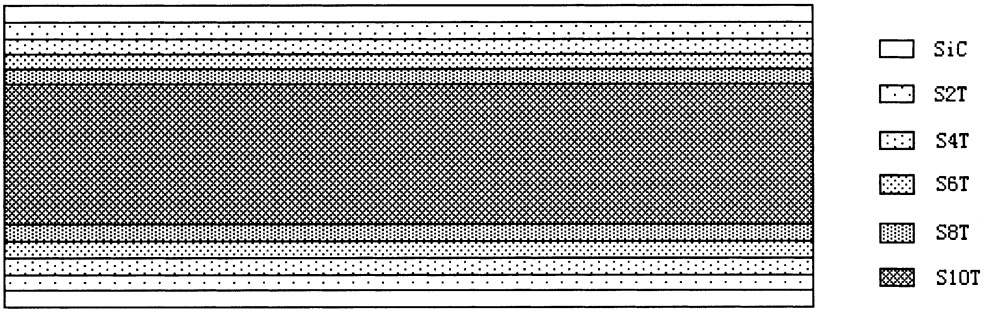

Fig. 3. The model of finally chosen symmetrical SiC/S10T structure (where S2T is SiC + 2 wt.% TiC, S4T is SiC + 4 wt.% TiC, and so forth).

for pyrolysis of the organic components at 700 °C, then hot-press sintered at 1850 °C, under 35 MPa pressure, for 30 min. The sintered materials were ~4 mm thick.

The relative density of the composite was calculated by Archimedes method. The ceramic then was cut to nominal 3×4×40 mm (width×thickness×length) specimens for mechanical testing by the three-point bend method. For comparison, layered pure SiC and S10T ceramics were fabricated by the same process. The surfaces of all of the specimens were ground and beveled carefully on a glass plate, using 32 μm and, then, 0.6 μm SiC powder. One side face of the SiC/S10T specimen was polished and observed by optical microscopy. The specimens for the fracture toughness test were single-edge notched on the layer plane, at a notch depth of about one-quarter of the specimen thickness, according to the detailed study of Lakshminarayanan et al.[17] The load was normal to the layer plane. The mechanical tests were conducted (Model 810, MTS Systems, Eden Prairie, MN) at a crosshead speed of 0.1 mm/min. Six specimens were tested for each value.

The stress conditions for the center of the surfaces of the layered SiC/S10T composites and a layered pure SiC fabricated by the same processes, which was selected as the standard sample, were tested using an X-ray stress analyzer (Model X-350A, Handan X-ray Institute, Handan, China). The X-ray diffraction method for measuring strains and stresses in crystalline solids has been well established.[18] In the past years, many studies on measurements of residual stress and strain in ceramic matrix composites by X-ray diffraction were done.[19–22] The method essentially uses the crystal lattice as an absolute strain gauge.

When an external load is applied to a crystal the lattice distorts and d changes. By comparing the measured d value in a distorted crystal with the strain free interplanar spacing d_0 the elastic strain (ε) can be determined

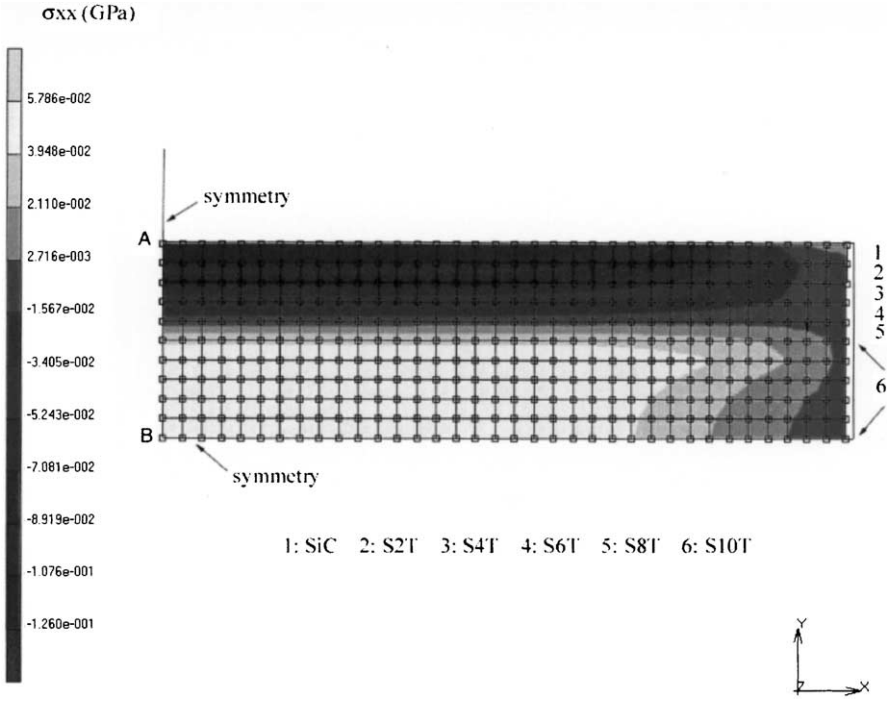

Fig. 4. The σ_{xx} distribution of one-quarter of the model in Fig. 3.

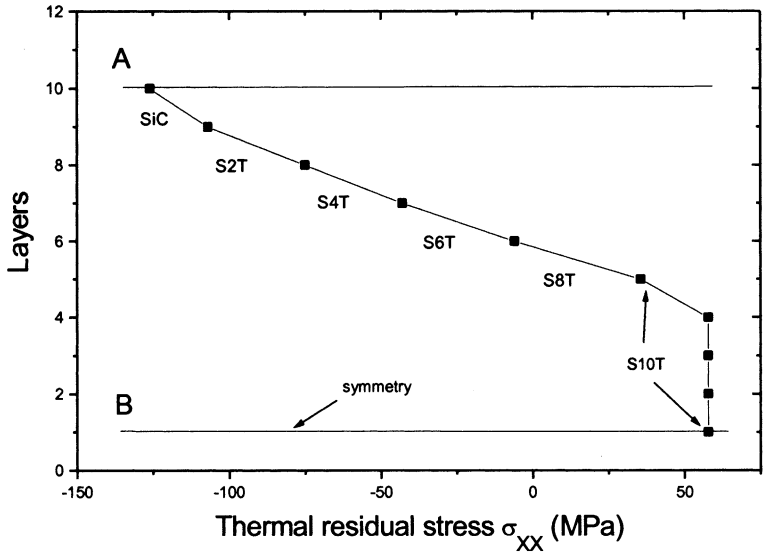

Fig. 5. The σ_{xx} distribution from point A to point B in Fig. 4.

$$\varepsilon = \frac{d - d_0}{d_0} = \cot\theta_0 \frac{\Delta(2\theta)}{2}$$

where 2θ is the angle between the incident and diffracted beams and λ the incident wavelength.

From the elastic strain, the corresponding stress can be calculated with the aid of elastic constants appropriate for the hkl plane used.

In this work, Cr K_α radiation was used as the X-ray source. The penetration depth of the X-ray was less than 10 μm. The (222) plane of SiC was selected as the diffraction crystal face. Biaxial stress on the surface of the specimens was assumed and the stress along the length direction of the specimens was tested. The widely recognized $\sin^2\psi$ method was used and measurements were made at two ψ values of 0 and 45°, corresponding to $\sin^2\psi$ values of 0 and 0.5. Firstly, the surface stress of the layered pure SiC ceramic was tested as a standard stress. Then the surface stress of the layered SiC/S10T ceramic was tested. The tested stress of the layered SiC/S10T ceramic minus the standard stress was regarded as the surface thermal residual stress of the layered SiC/S10T ceramic.

The fracture surface and the titanium content on the polished face were studied by electron probe micro-analysis (EPMA; Model 8705, Shimadzu Corp., Kyoto, Japan).

4. Results and discussion

Fig. 6 shows the optical microstructure of the SiC/S10T layered ceramic. After sintering, the interfaces between the different SiC+TiC layers disappeared completely. The light phase in Fig. 6(c) is TiC particles. The titanium content from the SiC to the S10T layer was detected qualitatively by EPMA spectrum analysis (Fig. 7), which revealed a gradual distribution of TiC from the surface to the inner layer.

The X-ray stress analysis results testified to a uniaxial compressive stress of 129 MPa at the center of the surface of the bending specimen. This value is very close to that from the FEM calculation, which showed a 126 MPa compressive stress at the center of the SiC layer (Fig. 5).

Table 2 lists the mechanical properties of the layered SiC, S10T and SiC/S10T ceramics. The pure S10T layered ceramic has higher fracture toughness, but a lower strength, than the SiC. The SiC/S10T layered ceramic shows excellent strength and toughness.

Theoretically, if the fracture of an SiC/S10T ceramic occurs from the SiC surface layer, the strength of the SiC/S10T layered ceramic should equal that of the pure layered SiC plus the calculated surface compressive residual stress. Table 3 shows that the tested strength of the SiC/S10T was very close to the theoretical value. This result indicates that surface residual stress was completely effective in strengthening the ceramic.

Fig. 8 shows fractographs of the present ceramics. The SiC grains in the pure SiC, S10T and SiC/S10T ceramics were fine, 1 μm. All of the ceramics fractured along the SiC grain boundaries. The fracture of the TiC particles, shown in Fig. 8(b), was caused by high tensile stress in the TiC particles, resulting from the large CTE mismatch between SiC and TiC. The ceramics were dense, with relative densities of 99%, according to density test results. The fracture toughness of the SiC/S10T was higher than that of the SiC and S10T. The increased toughness was derived from a decrease in the crack-driving force caused by surface compressive stress. In fact, the intrinsic fracture toughness of the layer materials did not improve. Thus, the phenomenon of the

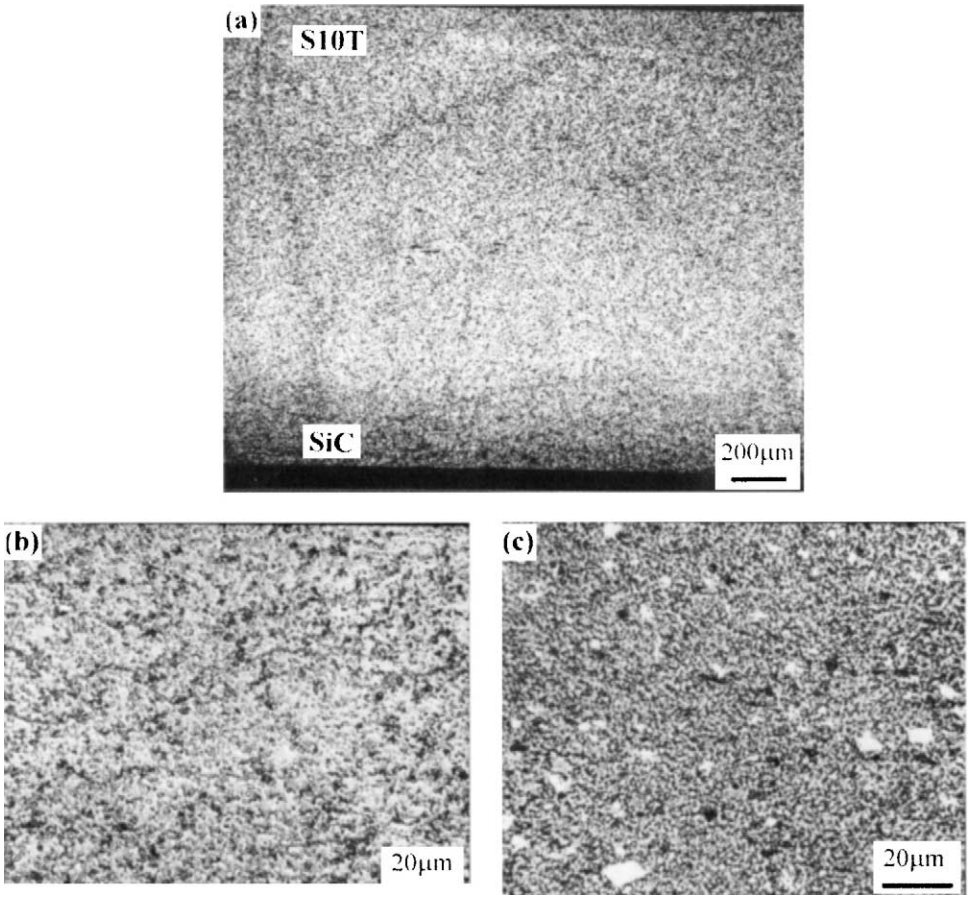

Fig. 6. The optical microstructure of the SiC/S10T layered ceramic: (a) low magnification, (b) SiC layer, high magnification, (c) S10T layer, high magnification.

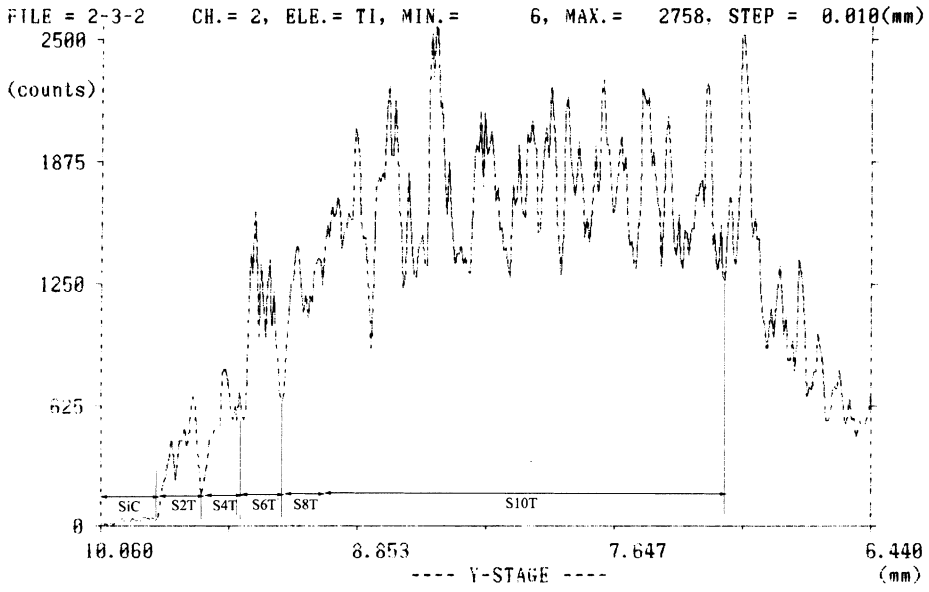

Fig. 7. Spectrum analysis of the titanium content from the SiC to the S10T layer.

Table 2
Mechanical properties of layered SiC, S10T and SiC/S10T ceramics

Materials	SiC	S10T	SiC/S10T
Bending strength (MPa)	714±86.3	663±38.7	834±73.1
Fracture toughness (MPa $\sqrt{m}$)	6.66±0.97	7.86±0.72	10.54±0.15

improvement of the fracture toughness of the SiC/S10T layered ceramic is only apparent. The tested toughness of the SiC/S10T was much more uniform than those of the monolithic SiC and S10T ceramics. This uniformity was caused by the crack-shielding effect of the surface compressive stress. Lakshminarayanan et al.[17] have proved that compression-strengthened ceramics have a

Table 3
Comparison of tested strength of SiC/S10T layered ceramic with theoretical

Strength of pure SiC (MPa)	Calculated residual stress on surface of SiC/S10T (MPa)	Tested residual stress on surface of SiC/S10T (MPa)	Theoretical strength of SiC/S10T (MPa)	Tested strength of SiC/S10T (Mpa)
714	−126	−129	840	834

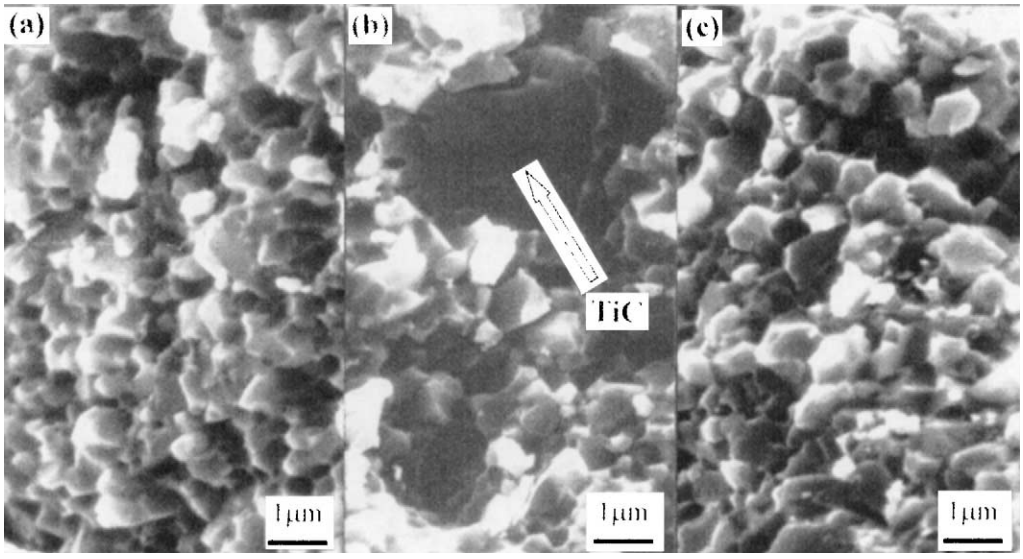

Fig. 8. Fractographs of the present ceramics: (a) the pure SiC, (b) the pure S10T, (c) the SiC/S10T.

strong tendency to trap surface cracks in the outer layer. Thus, the crack tolerance of a structure with an edge crack is enhanced, and the flaw sensitivity of the fracture toughness of a layered structure with surface compressive stress is decreased.

5. Conclusions

The finite element method (FEM) is suitable for designing layered ceramics with gradual thermal residual stress. A symmetrical SiC/S10T layered ceramic designed by FEM can be fabricated by means of aqueous tape casting, stacking, pyrolysis of organic components, and hot-press sintering. The surface stress conditions of SiC/S10T layered ceramics tested by X-ray stress analysis are close to the FEM calculation results, and the tested strength of the SiC/S10T is very close to the theoretical value.

Surface compressive residual stress is the main strengthening mechanism. Layered SiC/S10T ceramics have a higher fracture toughness than pure SiC and S10T ceramics, as a result of the decrease in crack-driving force caused by surface compressive stress. However, this phenomenon of the improvement of the fracture toughness is only apparent. The tested toughness of the layered SiC/S10T is much more uniform than that of monolithic ceramics. The uniformity is caused by the crack-shielding effect of the surface compressive residual stress.

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