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Influence of Interlayer Materials on Mechanical Properties of SiC Matrix Laminated Ceramic Composites

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SiC matrix laminated ceramic composites with various interlayer materials, such as high-melting metal tungsten, SiC without sintering aids, BN-Al_2O_3 and BN-Al_2O_3-SiC, were investigated in this study to determine the influences of interlayer on mechanical properties and fracture mechanisms of laminated composites. Gelcasting and hot-pressure sintering technologies were used to obtain the testing materials. The results revealed that the metal interlayer could not take the effect of deflecting crack because some metallic carbide and metallic silicide were formed by the reaction between metal and SiC during sintering process. The pure SiC interlayer disappeared after sintering and the composite became a block-like material due to the infiltration of liquid sintering aids from matrix. For BN-Al_2O_3 interlayer, the fracture toughness of laminated composite improved substantially, but meanwhile the flexural strength of composite decreased significantly because of low strength of the interlayer material. When adding some SiC in BN-Al_2O_3, the strength of interlayer material and the bond strength between interlayer and matrix layer enhanced, so that the fracture toughness and the flexural strength of the laminated composite increased simultaneously compared to the composite with BN-Al_2O_3 interlayer. It was found from the observation of crack propagation path that the crack could be deflected effectively in the laminated composite with BN-Al_2O_3-SiC interlayer.

Key words: laminated ceramic composites, interlayer, silicon carbide, mechanical properties, fracture mechanism

1. INTRODUCTION

Silicon carbide have now been recognized as potential candidates for high temperature structural applications in advanced heat engine parts and heat exchangers because of its high mechanical strength, high chemical stability and good thermal conductivity at elevated temperatures [1,2]. However, the widespread application of SiC ceramic is limited by its poor fracture toughness. Various kinds of SiC matrix composites, such as composites reinforced by second phase particle, whisker or fiber etc, have been studied to improve the toughness. Although the fiber reinforced SiC composites have been demonstrated to be an effective way, its high cost is also a big barrier for application. Laminated ceramic composites have attracted increasing attention since Clegg et al. [3, 4] developed a cheap and effective process to produce multilayered SiC ceramics with high toughness in 1990s. It has been known that the brittleness of ceramics can be improved by introducing weak interlayers, which may deflect a growing crack and thus prevent catastrophic failure. Most of the researches on laminated ceramic composites with weak interlayer, until now, were focused on SiC/C and Si_3N_4/BN systems [5-8]. High toughness, work-of-fracture and reliability were exhibited in the two composites systems due to the presence of weak C or BN interlayers. Therefore, the interlayers play a key role in these materials. In this study, SiC matrix laminated ceramic composites with various interlayer materials, such as high-melting metal tungsten, SiC without sintering aids, BN-Al_2O_3 and BN-Al_2O_3-SiC, were investigated to determine the influences of interlayer on mechanical properties and fracture mechanisms of laminated composites.

2. EXPERIMENTAL PROCEDURES

2.1 Preparation of testing materials

Silicon carbide powder, manufactured by Zhengzhou Dongfang Machinery Company, China, was used in this study. The mean particle size of the powder is 0.7 μm, and its chemical composition is given in Table 1.

Table I. Chemical composition (wt%) of SiC powder

SiC	Free carbon	Free silicon	SiO_2	Fe_2O_3
99.3	0.11	0.071	0.42	0.013

The sintering additives were 7 wt% Y_2O_3 (99.99%, Jiangxi Huaxin Group, China) and 3 wt% Al_2O_3 (99.9%, Zhangjiakou Special Ceramics Plant, China). Aqueous Gel Casting (AGC) technology [9] was used to prepare the SiC matrix green layer with a thickness of 0.4mm. For laminated composite with metal interlayer, metal tungsten powder (Yinchuan Refractory Metal Company, China) was coated on the surface of SiC green layer by spray technique. After stacking the green layers into a graphite die, hot pressing was conducted for 1h at 1800 ℃ under a pressure of 25MPa in Ar atmosphere. For laminated composite with different ceramics interlayers which include pure SiC, BN-Al_2O_3 and BN-Al_2O_3-SiC, both surfaces of each matrix green layer were deposited with the interlayer materials slurry by a dip-coating method, and then the laminated composites were prepared by the same hot-pressing technique but in N_2 atmosphere. The SiC matrix block material was also achieved by hot-pressing for the purpose of comparison.

2.2 Experimental method

The laminated composites and SiC block material were cut and ground into rectangular bars of 3mm (height) by 4mm (width) by 36mm (length) and 5mm (height) by 2.5mm (width) by 25mm (length) for strength and toughness test, respectively. Mechanical strength was evaluated by three-point bending test with a support distance of 30mm and the cross-head speed was 0.5mm/min. The tensile surfaces were polished and the edges were chamfered. Fracture toughness was measured by four-point single edge notched beam (SENB) technique with a 10mm inner span and a 20mm outer span at the cross-head speed of 0.05mm/min. The notch depth and width were about 2.5mm and 0.25mm, respectively. Strength and toughness measurements were conducted in a MTS-810 universal testing machine with the layer plane of samples perpendicular to the loading direction. The load–displacement response was recorded with a computerized data-acquisition system. The microstructure of fracture surface was observed by AMRAY-1000B scanning electron microscope. The crystalline phases in SiC/W composite were identified by X-ray diffraction (XRD, Rigaku D/MAX2000).

3. RESULTS AND DISCUSSION

3.1 Mechanical properties of testing material

The flexural strength and fracture toughness of testing materials are shown in Table II. It can be seen from the table that the SiC matrix block material possesses good mechanical properties. The flexural strength and fracture toughness of the composite with high-melting metal tungsten interlayer decreased, and the strength enhanced and toughness dropped for the composite with pure SiC interlayer comparing to the SiC matrix. When $BN-Al_2O_3$ interlayer was introduced, a dramatic change happened. The toughness of the composite was about three times higher than that of the SiC matrix, meanwhile, the strength of the composite reduced markedly. By adding SiC in $BN-Al_2O_3$ as interlayer material, the strength and toughness of $SiC/BN-Al_2O_3-SiC$ laminated composite increased simultaneously comparing with the $SiC/BN-Al_2O_3$ composite. Therefore, the mechanical properties of laminated ceramic composites are related strongly with the kind of interlayer materials.

Table II. Mechanical properties of the testing materials

Materials	Flexural strength MPa	Fracture toughness $MPam^{1/2}$
SiC	661	7.5
SiC/W	560	7.3
SiC/SiC	761	6.7
$SiC/BN-Al_2O_3$	155	20.0
$SiC/BN-Al_2O_3-SiC$	277	23.8

3.2 SiC matrix block material

As shown in Table I, the SiC raw material is a high purity powder. The microstructure of the SiC matrix block material on fracture surface is shown in Fig.1. It can be seen from the figure that the average grain size of the SiC ceramics was about 1.5 μm, and the specimen shows an intracrystalline fracture surface. Some grains were pulled out, and a certain undulated fracture surface could also be seen from the figure, so the SiC ceramics exhibits good mechanical properties.

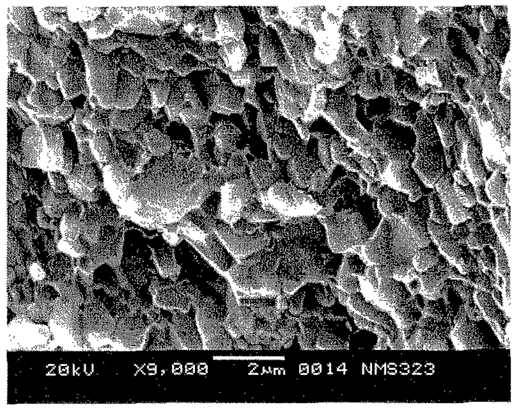

Fig.1 SEM micrograph of SiC matrix block material on fracture surface.

3.3 Composite with tungsten interlayer

Metal tungsten is a refractory metal with very high melting point (3683 K), low thermal expansion coefficient (5.5×10^{-6} K^{-1}) and good ductility. Metal tungsten seems an appropriate interlayer metal for making SiC based composites judging from its physical properties. Whereas, it was demonstrated by the present work that SiC/W composite could not toughen the SiC matrix ceramic. The XRD pattern of the composite is shown in Fig.2. It can be seen that not ductile metal tungsten but brittle tungsten carbide (WC) and tungsten silicide (W_5Si_3) existed in the composite. Therefore, the composite was weakened actually, and its strength and toughness are lower than that of the SiC matrix.

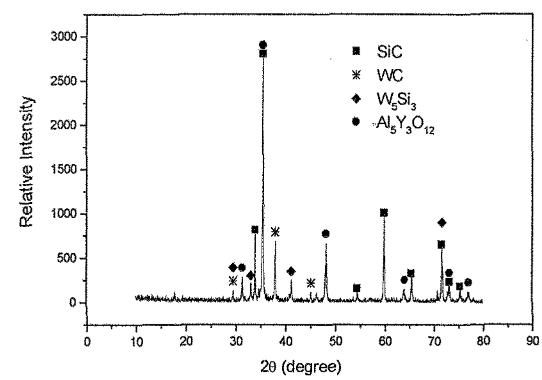

Fig.2 X-ray diffraction pattern of SiC/W composite

In SiC-W ternary system, chemical reaction between SiC and W is easy to proceed, and the stability of phases is a function of temperature [10, 11]. At lower temperatures, nominally below 970 K, following reaction will proceed:

$$2SiC+3W \rightarrow 2WC+WSi_2 \quad (1)$$

Above this temperature (up to 2140 K), WSi_2 is no longer stable, and W_5Si_3 will be produced according to the following reactions:

$$3SiC + 8W \rightarrow 3WC + W_5Si_3 \quad (2)$$

$$WSi_2 + SiC + 5W \rightarrow WC + W_5Si_3 \quad (3)$$

The sintering temperature of the composite was 2073K, thus the reaction products are WC and W_5Si_3 as shown in Fig.2. Yttrium aluminium garnet ($Y_3Al_5O_{12}$ or YAG), the product form sintering additives Y_2O_3 and Al_2O_3, can also be seen from the figure.

3.4 Composite with pure SiC interlayer

Pure SiC powder could not be well sintered by hot-pressing without sintering additives, so it was considered at the beginning of this study that perhaps the pure SiC is a suitable interlayer material to separate the matrix layers. Fig.3 shows the macro morphology of fracture surface in SiC/SiC composite by SEM. It can be seen that there is no crack deflection formed in the material in spite of some surface fluctuation. The fracture surface of SiC/SiC composite looks like a block ceramic. When the composite was hot pressed at elevated temperature, the liquid of the sintering additives could permeate easily from matrix layer to interlayer, and pure SiC interlayer was completely sintered together with the matrix layer. Therefore, pure SiC can not exist and form a real interlayer in the material after sintering, and the SiC/SiC material, in fact, is not a laminated composite.

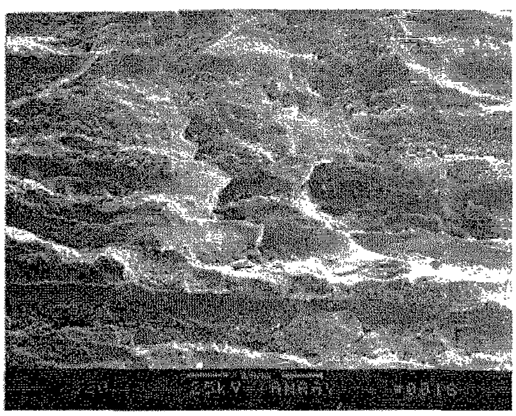

Fig.3 Macro morphology of fracture surface in SiC/SiC composite.

3.5 Composite with $BN\text{-}Al_2O_3$ interlayer

It has been identified that $BN\text{-}Al_2O_3$ is a practicable interlayer material for Si_3N_4 matrix laminated composite [7, 8]. In this study, $BN\text{-}Al_2O_3$ material was used as the interlayer of SiC matrix composite. The significant effect of $BN\text{-}Al_2O_3$ interlayer on the toughness has been revealed in Table II. The fracture path of $SiC/BN\text{-}Al_2O_3$ composite is shown in Fig.4. It can be seen that the through-thickness crack is deflected distinctly by the $BN\text{-}Al_2O_3$ interlayer, and fracture surface presents a step-like appearance. During the fracture process, a through-thickness crack will be arrested when it moves across the a matrix layer. It will run into the adjacent $BN\text{-}Al_2O_3$ interlayer because the three-dimension stresses on the crack-tip are relaxed by the weak interlayer. The crack propagates in the interlayer along the layer plane. A delamination occurs in the interlayer and the through-thickness crack is deflected. As the displacement continues, the tensile stress on the next matrix layer increases constantly until the uncracked portion of the specimen cannot support the applied load. Then, a new through-thickness crack forms in the adjacent matrix layer and is deflected by the next interlayer again. This process is repeated until the complete fracture of specimen. Therefore, the fracture of composite occurs gradually but not catastrophically, and its fracture toughness increases markedly comparing with the matrix ceramic material.

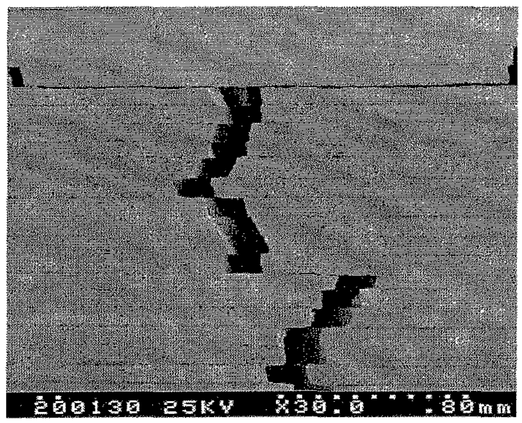

Fig.4 Fracture path of $SiC/BN\text{-}Al_2O_3$ composite.

On the other hand, the $BN\text{-}Al_2O_3$ interlayer is very weak in mechanical strength, and it works as a two-dimension flaw in the laminated composite. Although the interlayer can play the deflection role, the interlayer is delaminated easily, and sometimes, the deflected crack propagated through the whole interlayer, as shown at the top of Fig.4. As a result, the flexural strength of laminated composite decreased remarkably.

3.6 Composite with $BN\text{-}Al_2O_3\text{-}SiC$ interlayer

When some SiC powder was added in the $BN\text{-}Al_2O_3$ interlayer, it can be seen in Table II that strength and toughness of the laminated composite are increased from 155 to 277MPa and 20.0 to 23.8MPam$^{1/2}$, respectively. The addition of SiC can improve the strength of the interlayer and the bond strength between interlayer and matrix layer. After the through-thickness crack is deflected, the delamination in the interlayer is more difficult, and more energy is consumed in the delamination process. As a result, the strength and toughness of the laminated composite increased simultaneously. It is obvious that the strength of the interlayer should be suitable, and too strong or too weak interlayer will be detrimental to the improvement of toughness of laminated composites. Fig.5 shows the typical crack propagation path and the corresponding load-displacement response in $SiC/BN\text{-}Al_2O_3\text{-}SiC$ laminated composite. It can be seen that the crack was deflected sufficiently. There are several peaks in the load-displacement plot, and the total fracture displacement of the laminated composite is increased significantly comparing with the matrix material. Therefore, the fracture toughness and fracture work of

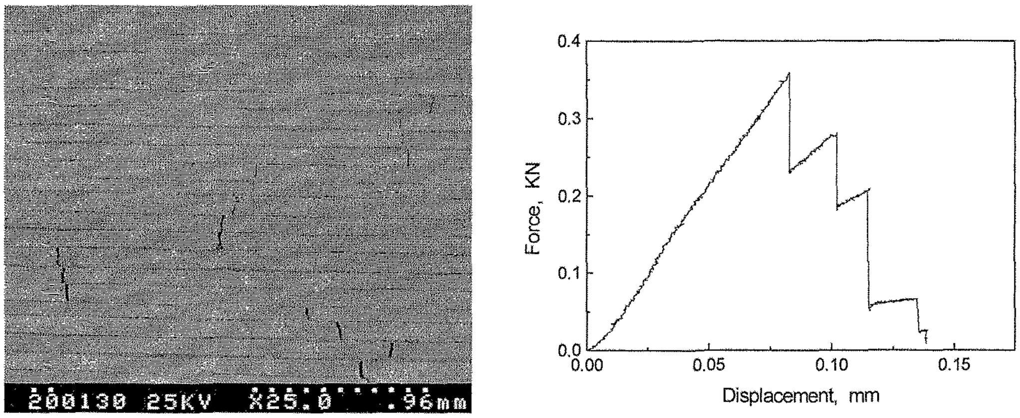

Fig.5 Fracture path and corresponding load-displacement response in SiC/BN-Al_2O_3-SiC laminated composite

laminated composite are improved essentially by its special structure.

4. CONCLUSIONS

The influences of various interlayers on mechanical properties of SiC matrix laminated ceramic composites were investigated in this paper. Gel-casting and hot-pressing were used to fabricate the testing materials. It is shown that the high-melting metal tungsten interlayer did not take the effect of deflecting crack because some metallic carbide and metallic silicide were formed by the reaction between metal tungsten and SiC during sintering process. The attempt of pure SiC interlayer is also not successful. The interlayer disappeared after sintering and the composite became a block-like material due to the infiltration of liquid sintering aids from matrix. The fracture toughness of laminated composite with BN-Al_2O_3 interlayer improved substantially because the crack can be deflected effectively, but its flexural strength decreased significantly due to the low strength of interlayer material. When the strength of BN-Al_2O_3 interlayer enhanced via adding in some SiC powder, the fracture toughness and flexural strength of laminated composite are increased from 20.0 to 23.8MPam$^{1/2}$ and 155 to 277MPa, respectively. Therefore, the mechanical properties of laminated ceramic composites are related strongly with the choice of interlayer materials.

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