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Ceramics International 30 (2004) 697–703

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Fracture behavior of laminated SiC composites

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Received 25 May 2003; received in revised form 19 June 2003; accepted 25 July 2003

Abstract

Experimental investigation of the flexural properties of SiC/C laminates had been conducted. Effect of the interfacial thickness and composition on the mechanical properties of SiC laminates was characterized. The diffusion of elements from adjacent SiC layers in the carbon-based interfacial layers was studied. Accordingly, the optimal thickness and composition of interfacial layers were determined experimentally.

Keywords: B. Composites; C. Mechanical properties; C. Fracture; D. SiC

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1. Introduction

Active research and development have been conducted to seek the application of ceramics as structural components; however, the lack of damage tolerance ability has been a critical problem. It was reported that continuous-fiber-reinforced ceramic composites (CFCC) are the most promising candidates and have shown a non-brittle response [1], but the fabrication of these advanced structural materials for high temperature applications is time-consuming and expensive. The concept of laminated composite for improved performance of brittle materials is well established [2]. Such structure is found in many biological hard tissues, such as mollusk shell [3,4] and teeth. Clegg and co-workers [5–8] have produced laminated SiC with graphite interface layers by rolling the dough like powder mixture into sheets, coating the sheets with graphite followed by laminating and sintering. These multilayer SiC composites showed stepped stress–strain behavior with the apparent toughness and work of fracture as $15\,\mathrm{MPa\,m^{1/2}}$ and $4625\,\mathrm{J\,m^{-2}}$, respectively. Multilayered ceramic materials with weak interface have been evaluated in other ceramic systems, such as SiC/SiC [9], Si_3N_4/BN [10–15] or Al_2O_3/$LaPO_4$ [16], etc. Recently, it was shown that laminated composites without weak interfaces also exhibited damage-tolerant behavior [9,17–20].

The individual layers of laminates with weak interface are usually fabricated by extrusion [5,6,8,21], slip casting [19], electrophoretic deposition [22], non-aqueous tape casting [10,12,14,15,17,18,20] and laminated object manufacturing [23], no paper had ever reported the using of aqueous tape casting process. As far as we know, the interface layer of laminates are prepared by coating [5,6,8,10,12], spraying [21], electrophoretic deposition [22], screening [14] and tape casting [12,15,17,18].

Understanding the fracture behavior of multilayered composites requires the study of interface crack propagation mechanism [24–29]. Phillipps et al. [7,30], Folsom [31,32], and others proposed models of the fracture behavior of ceramic laminates in bending test. The reliability and frictional energy dissipation in laminates were also investigated [33–35].

Phillipps et al. proposed that the specimen size, including the beam length, the interfacial toughness, the lamina thickness will play important roles on the apparent toughness, work of fracture of laminated composites [8]. In the present paper, SiC/C laminated composites were prepared through aqueous tape casting and lamination process. The properties of the thin C–SiC interfacial layers were adjusted by varying the composition of the C through the addition of SiC. Influence of the composition and thickness of interfacial layers on the mechanical properties of SiC multilayers was studied. The microstructure and fracture behavior were investigated too.

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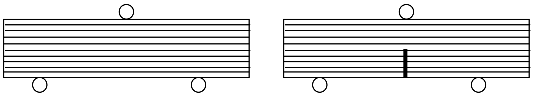
doi:10.1016/j.ceramint.2003.07.016



(a) (b)

Fig. 1. Schematic illustration of (a) the specimen orientations for three points bending, (b) a notched bend bar showing the notch orientation with respect to the layered structure.

2. Processing, mechanical testing and characterization methods

2.1. Lamination process

Silicon carbide sheets were prepared using commercial SiC powders (FCP-15, Norton com.) as starting materi-

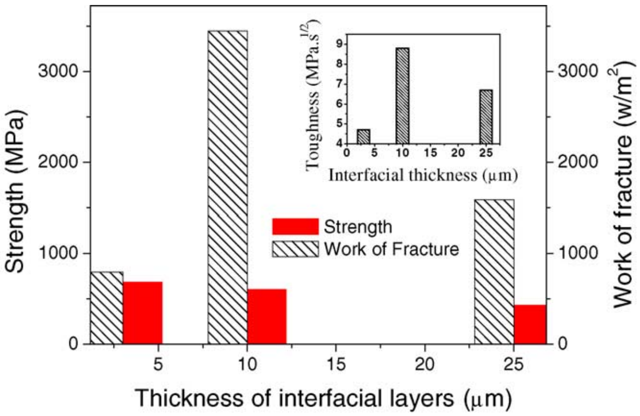
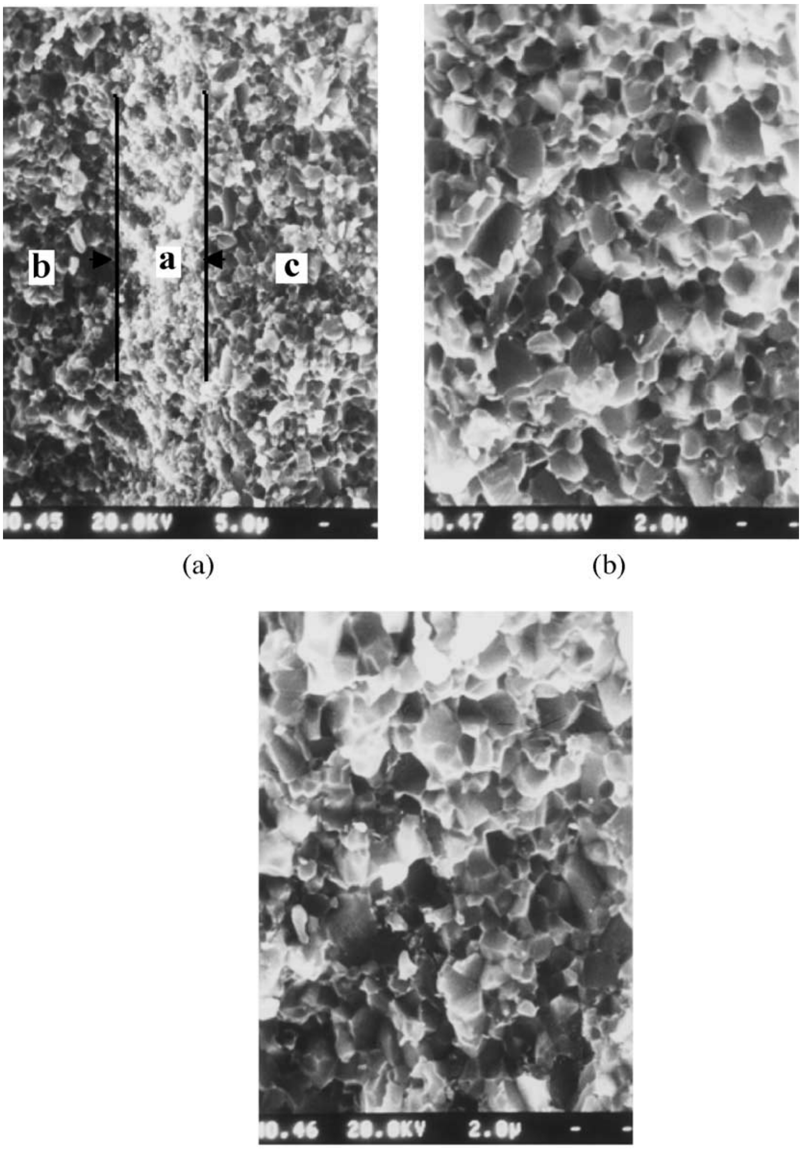


Fig. 2. Influence of the thickness of interfacial layers on the mechanical properties of SiC laminates.



(a) (b)

(c)

Fig. 3. Fractured surface of the laminated SiC composites (a) interfacial layer, (b) left layer, (c) right layer.

als and Y_2O_3 (2.85 wt.%) and Al_2O_3 (2.15%) as sintering additives. Details concerning the aqueous tape casting process could be obtained from previous published papers [36,37]. The thickness of the SiC green tapes was adjusted to ∼150 μm. To introduce weak interface between SiC layers, thin C–SiC sheets were also prepared by tape casting process using doctor blade equipment. The C powders (Optical pure, Shanghai Carbon Element Factory) were firstly dispersed in deionized water followed by the addition of SiC powders with controlled composition. PVA1788 was selected as binder and glycerol as plasticizer. The thickness of C sheets was adjusted to ∼30 μm. Much thinner layers were also prepared by the so-called “screen printing” process. The surface of each SiC sheets was coated by passing the aforementioned C slurries through a 200-mesh screen. The thickness of the interfacial layers was varied in the 5–20 μm range through the adjustment of solid content of the said slurry. After coating, the sheets were dried and stacked in the repeating sequence of SiC and C–SiC layers. Subsequently, organic additives were removed by heating to 800 °C in a flowing argon atmosphere. A slow heating rate was selected to minimize bloating and cracking during pyrolysis, which might result in distortion of the layers. After pyrolysis, the billets were placed in a BN-coated graphite die. Consolidation was performed by hot pressing at 35 MPa under an argon atmosphere at temperature of 1850 °C for 0.5 h.

2.2. Mechanical and microstructure evaluation of laminated composites

Specimens for flexural test were cut and ground into rectangular bars. A schematic illustration of bending test process for laminated SiC composites is shown in Fig. 1. Three-point bending strength was determined at room temperature on five to six 3 mm × 4 mm × 36 mm bars with a span of 30 mm and a cross head speed of 0.5 mm·min^{-1}. Apparent toughness was measured by the single-edge-notched-beam (SENB) method on test bars of 4 mm high, 3 mm wide and 36 mm long by a three-point bend using a span of 30 mm and a cross head speed of 0.05 mm·min^{-1}. A straight notch with fine diamond saw was introduced with a depth at about 2 mm. Work of fracture was obtained by dividing the area under the load–displacement curve by twice the cross-section area of the sample [38].

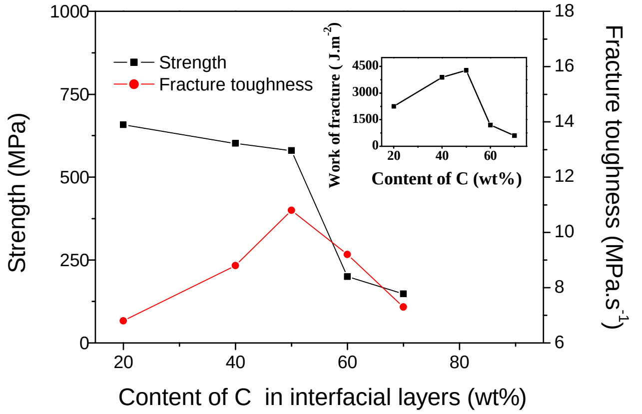


Fig. 4. Influence of interfacial composition on the mechanical properties of SiC laminates.

2.3. Microstructure characterization

Microstructure characterization was performed by optical microscopy and SEM. Energy dispersive X-ray (EDX) spectra were obtained across the interfacial layer to determine the diffusion of elements from adjacent layers. The crack pattern of fractured surface was also characterized.

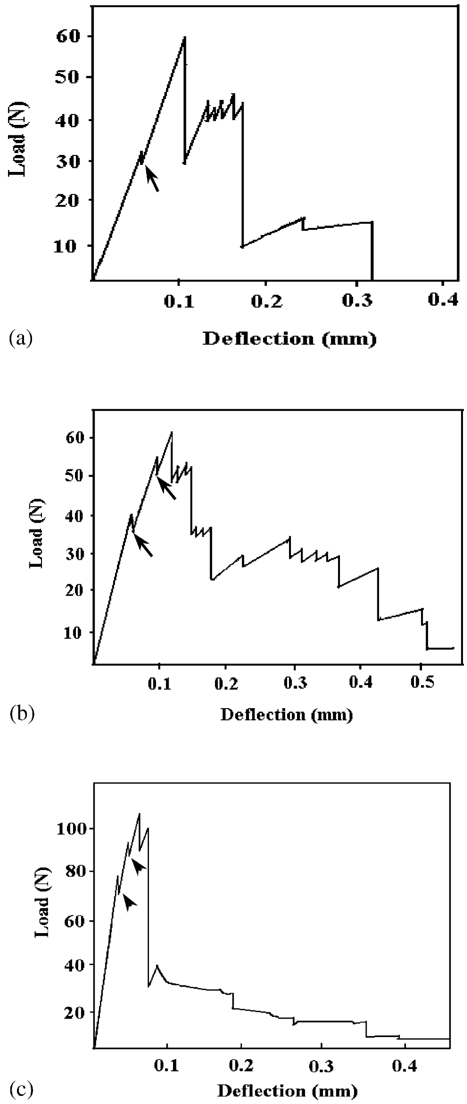


Fig. 5. Load–deflection curves of SiC laminates (a) SiC30-C70, (b) SiC40-C60, (c) SiC50-C50.

3. Results and discussion

3.1. Interfacial thickness

Effects of thickness of interfacial layers on the strength, apparent toughness and work of fracture of SiC samples are shown in Fig. 2.

The strength of SiC samples shows an increase trend with the decrease of the thickness of interfacial layers; while the apparent toughness and work of fracture exhibit a different character (see Fig. 2). When the interfacial thickness is ∼3 μm, SiC laminates exhibit a catastrophic fracture behavior; on the other hand, when the interfacial thickness is higher than 25 μm, graceful failure occurs, the mechanical properties of SiC laminates drop altogether. An optimal interfacial thickness is shown to be around 10 μm (see Fig. 2).

The microstructure of fractured surface of SiC samples is shown in Fig. 3. The belt between parallel lines is the interfacial layer, see Fig. 3(a), the left side (marked as "b") and right side (marked as "c") are shown in Fig. 3(b) and (c), respectively. The SiC layers (left side and right side) are well densified. However, it is difficult to detect exactly the interface between the interfacial layers and the adjacent SiC layers. This may be due to the diffusion of elements from SiC layers, which will be discussed later.

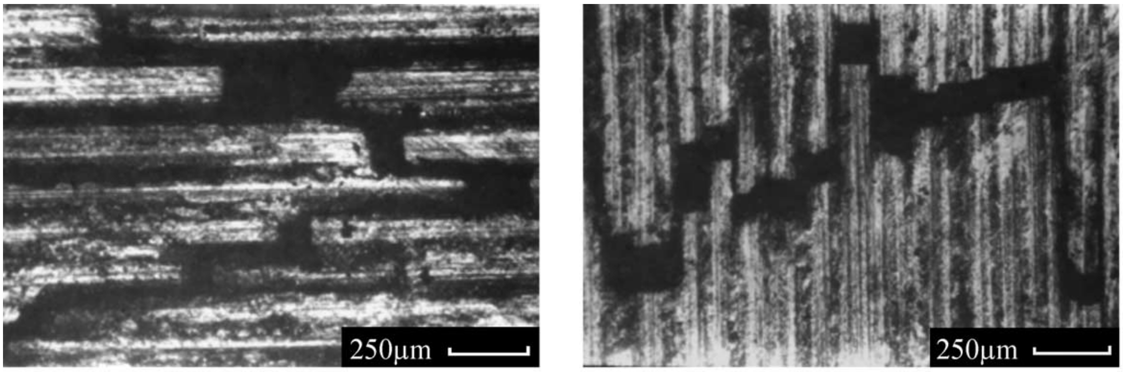
Table 1
Mechanical properties of interfacial layers

Sample	SiC30-C70	SiC50-C50	SiC60-C40
Strength (MPa)	7.33	15.67	19.33
Fracture toughness (MPa m$^{1/2}$)	0.30	–	0.34
Work of fracture (J m^{-2})	10	33.3	35.4

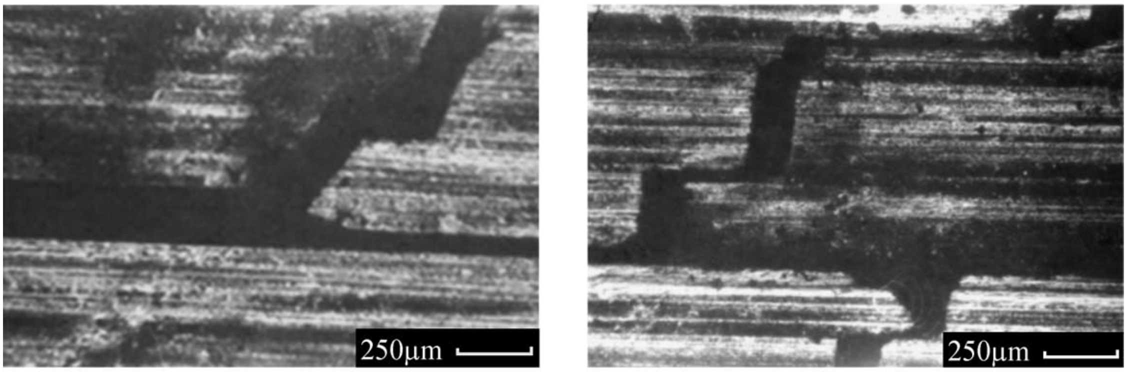
3.2. Interfacial composition and mechanical properties

The properties of interfacial layers are adjusted by controlling their composition. Effects of interfacial composition on the properties of SiC laminates are shown in Fig. 4. The C–SiC bulk samples with the same composition as interfacial layers were also sintered under the same condition for comparison. The mechanical properties of these C–SiC bulk samples are shown in Table 1.

As shown in Table 1, the addition of SiC powders has very limited effects on the mechanical properties of C–SiC bulk samples. However, as shown in Fig. 4, the mechanical properties of SiC laminates exhibit a considerable dependence on the composition of interfacial layers: the flexural strength decreases constantly with the increase of C in interfacial layers, similar to that of Si_3N_4 samples [13,14]. The fracture toughness and work of fracture first show an increase



(a) SiC40-C60 (b) SiC40-C60

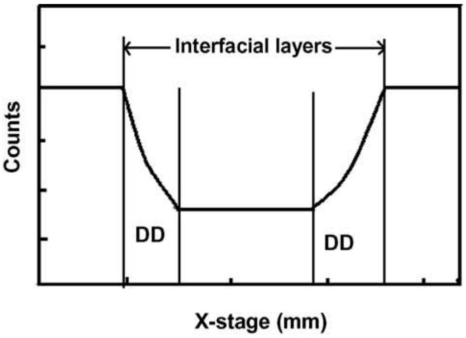


(c) SiC50-C50 (d) SiC60-C40

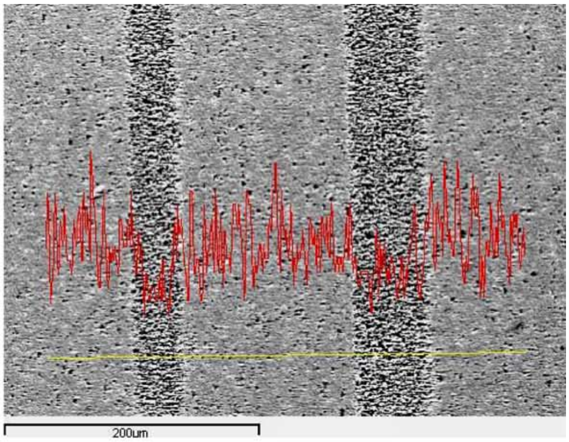
Fig. 6. Optical micrographs showing the side surface of flexural specimens containing (a) 40 vol.%, (b) 40 vol.%, (c) 50 vol.%, (d) 60 vol.% SiC in the interfacial layers (after testing).

trend with the increase of content of C in interfacial layers, up to approximately 50 vol.%, and decrease thereafter. For the interfacial composition of 50 vol.% SiC + 50 vol.% C, the strength, work of fracture and fracture toughness was 580 MPa, 4282 J·m^{-2}, and $K_{IC} = 10.8$ MPa·m$^{1/2}$, respectively. This strong dependence on interfacial composition cannot be well correlated with the results shown in Table 1. The only possible reason is the diffusion of elements (Al, Y, Si, etc.) from adjacent SiC layers, as discussed later.

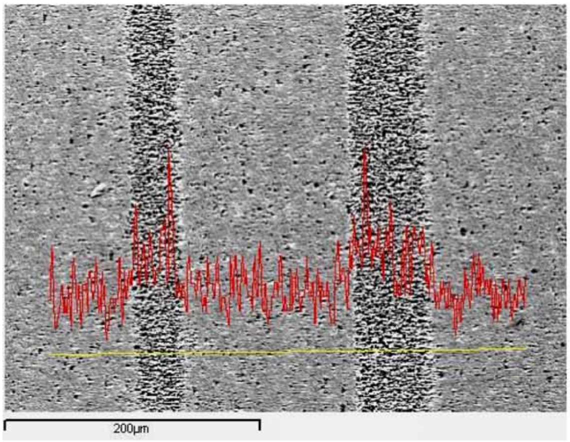
The nominal stress on the tensile surface is plotted versus cross head deflection of notched specimens with varied C



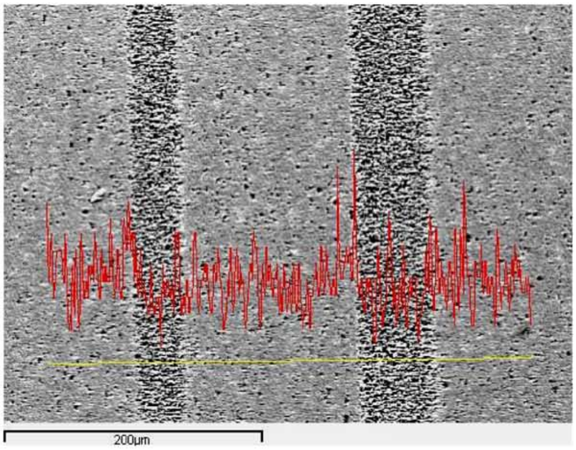
(a)



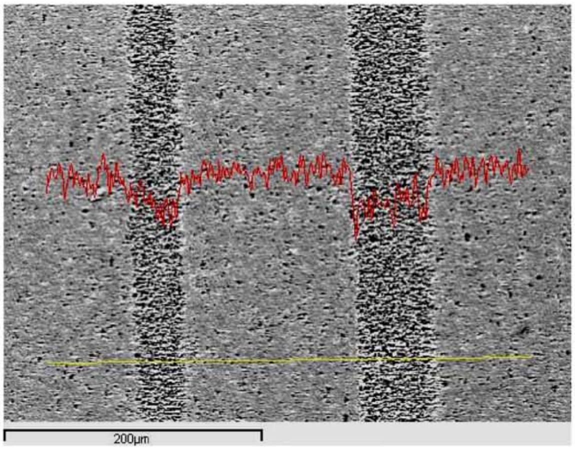
(b) Al



(c) C



(d) Y



(e) Si

Fig. 7. Diffusion of Al_2O_3, and Y_2O_3 in interfacial layers (a) schematic illustration, (b), (c), (d), (e) EDX characterization.

content (Fig. 5). Cracks are deflected in the interfacial layers until the SiC content is increased to 80 vol.%.

The micrographs of SiC laminates are shown in Fig. 6. It is observed that some cracks in SiC layers initiate from the growth of defects within the SiC lamina, while other cracks from the growth of the delaminaton cracks propagating along the interfacial layers. The latter would eliminate the fracture toughness and WOF although graceful failure may still occur. During the failure process, frictional sliding at the debonded interface may also take effect [35], which cannot be detected from the optical micrographs (Fig. 6).

3.3. *Diffusion effect*

As proposed by Cook and Gordon [2] and Clegg et al. [5,6], the weak interfacial layers were deliberately introduced as a toughening mechanism for deflecting growing cracks. Studies on the fracture behavior of laminated composites also showed that the critical ratio of strength and apparent toughness between interfacial layers and the adjacent matrix layers for crack deflection were relatively low [2,7,8,25,26,28,31,32]. So it can be proposed that the interfacial layers should be as thin as possible as long as these cracks could be deflected. However, specimens with the interfacial thickness as $\sim 3\,\mu\text{m}$ did not show the graceful failure behavior (see Fig. 2). This might be due to the diffusion effects. During sintering or other high temperature process, elements in adjacent layers will diffuse to the interfacial layers due to the composition difference. In this case, the residual stress will not be a significant factor due to the very limited strength and thickness of interfacial layers.

To observe clearly the diffusion phenomena, samples with thick interfacial layers are characterized by EDX, see Fig. 7(a)–(e). A schematic illustration of the diffusion phenomena is shown in Fig. 7(a).

As shown in Fig. 7, the distribution of Al, Y, Si, etc. elements across the interfacial layers exhibits a similar trend, suggesting some diffusion of these elements from adjacent SiC layers. However, due to the low content of sintering additives (Al_2O_3 and Y_2O_3), the EDX curves show a high undulation near the interface. It is difficult to characterize exactly the diffusion distance of elements (Al, Y, Si, etc.) in interfacial layers. Based on estimation from Fig. 7, the diffusion distance should be less than $10\,\mu\text{m}$. Assuming $10\,\mu\text{m}$ as the distance for diffusion of Si, Al, and Y into the interfacial layers, then the proper interfacial layer thickness should be about $20\,\mu\text{m}$. For thicker interfacial layers ($\gg 20\,\mu\text{m}$), such diffusion has negligible effect on the mechanical properties of interfacial layers, SiC laminates might exhibit a pronounced interfacial delamination failure behavior though the mechanical properties are largely reduced. In this case, the mechanical properties of SiC laminates is irrelevant to the thickness of the interfacial layers. On the other hand, for very thin interfacial layers ($\ll 20\,\mu\text{m}$), the diffusion of elements (Al, Y, Si, etc.) will make the interfacial layers "strong," therefore, the failure of SiC laminates may experience a transition from graceful to catastrophic mode: delamination may still occur, but the failure of the subsequent lamina will be determined by the delamination cracks across the interface. In this case, the mechanical properties of SiC laminates are irrelevant to the thickness of the interfacial layers too. Further reduction in the thickness of interfacial layers may result in a monolithic fracture mode. Clegg [6] related this kind of behavior to the existence of gaps in the interfacial layers. However, this explanation is not applicable to our case.

The influence of interfacial composition on the mechanical properties of SiC laminates can also be well correlated to the interfacial diffusion mechanism. The higher the content of C in interfacial layers, the weaker the interfacial layers after diffusion, consequently, the lower the strength. This explanation is also applicable to the fracture toughness and work of fracture.

4. Conclusion

The mechanical properties of SiC laminates depend on the thickness and composition of interfacial layers. For the interfacial composition of 50 vol.% SiC + 50 vol.% C, the strength, work of fracture and fracture toughness was 580 MPa, $4282\,\text{J}\cdot\text{m}^{-2}$, and $K_{IC} = 10.8\,\text{MPa}\cdot\text{m}^{1/2}$, respectively. Based on SEM and EDX observation, this dependence is mainly due to the diffusion of elements (Al, Y, Si, etc.) from adjacent SiC layers.

Acknowledgements

The authors were grateful to the Science and Technology Committee of Shanghai for providing support under the contract number 02DJ14065.

References

[1] A.G. Evans, Perspective on the development of high-toughness ceramics, J. Am. Ceram. Soc. 73 (2) (1990) 187–206.
[2] J. Cook, J.E. Gordon, A mechanism for the control of crack propagation in all-brittle materials, Proc. R. Soc. Lond. A282 (1964) 508–520.
[3] V.J. Laraia, A.H. Heuer, Novel composite microstructure and mechanical behavior of mollusk shell, J. Am. Ceram. Soc. 72 (11) (1989) 2177–2179.
[4] J.D. Currey, A.J. Kohn, Fracture in the crossed-lamellar structure of conus shells, J. Mater. Sci. 11 (1976) 1615–1623.
[5] W.J. Clegg, K. Kendall, N. McAlford, T.W. Button, J.D. Birchall, A simple way to make tough ceramics, Nature (London) 347 (1990) 455–457.
[6] W.J. Clegg, The fabrication and failure of laminar ceramic composites, Acta Metall. Mater. 40 (11) (1992) 3085–3093.
[7] A.J. Phillipps, W.J. Clegg, T.W. Clyne, Fracture behavior of ceramic laminates in bending-I. Modeling of crack propagation, Acta. Metall. Mater. 41 (3) (1993) 805–817.
[8] A.J. Phillipps, W.J. Clegg, T.W. Clyne, The correlation of interfacial and macroscopic toughness in SiC laminates, Composites 24 (2) (1993) 166–176.

[9] F. Rebillat, J. Lamon, R. Naslain, Properties of multilayered interphases in SiC/SiC chemical-vapor infiltrated composites with “weak” and “strong” interfaces, J. Am. Ceram. Soc. 81 (9) (1998) 2315–2326.

[10] H. Liu, S.M. Hsu, Fracture behavior of multiplayer silicon nitride/boron nitride ceramics, J. Am. Ceram. Soc. 79 (9) (1996) 2452–2457.

[11] E.H. Lutz, M.V. Swain, Fracture toughness and thermal shock behavior of silicon nitride–boron nitride ceramics, J. Am. Ceram. Soc. 75 (1) (1992) 67–70.

[12] Y. Shigegaki, Processing of a novel multilayered silicon nitride, J. Am. Ceram. Soc. 79 (8) (1996) 2197–2200.

[13] D. Kovar, M.D. Thouless, J.W. Halloran, Crack deflection and propagation in layered silicon nitride/boron nitride ceramics, J. Am. Ceram. Soc. 81 (4) (1998) 1004–1012.

[14] Z. Lenčéš, K. Hirao, M.E. Brito, M. Toriyama, S. Kanzaki, Layered silicon nitride-based composites with discontinuous boron nitride interlayers, J. Am. Ceram. Soc. 83 (10) (2000) 2503–2508.

[15] T. Ohji, Y. Shigegaki, T. Miyajima, S. Kanzaki, Fracture resistance behavior of multilayered silicon nitride, J. Am. Ceram. Soc. 80 (4) (1997) 991–994.

[16] P.E.D. Morgan, D.B. Marshall, Ceramic composites of monazite and alumina, J. Am. Ceram. Soc. 78 (6) (1995) 1553–1563.

[17] D.-H. Kuo, W.M. Kriven, A strong and damage-tolerant oxide laminate, J. Am. Ceram. Soc. 80 (9) (1997) 2421–2424.

[18] D.-H. Kuo, W.M. Kriven, Fracture of multilayer oxide composites, Mater. Sci. Eng. A A241 (1998) 241–250.

[19] A.J. Sanchez-Herencia, C. Pascual, J. He, F.F. Lange, ZrO_2/ZrO_2 layered composites for crack bifurcation, J. Am. Ceram. Soc. 82 (6) (1999) 1512–1518.

[20] H. Liu, B.R. Lawn, S.M. Hsu, Hertzian contact response of tailored silicon nitride multilayers, J. Am. Ceram. Soc. 79 (4) (1996) 1009–1014.

[21] J. She, T. Inoun, K. Ueno, Multilayer Al_2O_3/SiC ceramics with improved mechanical behavior, J. Eur. Ceram. Soc. 20 (2000) 1771–1775.

[22] L.J. Vandeperre, O.O. Van Der Biest, Graceful failure of laminated ceramic tubes produced by electrophoretic deposition, J. Eur. Ceram. Soc. 18 (1998) 1915–1921.

[23] D. Klosterman, R. Chartoff, G. Graves, N. Osborne, B. Priore, Interfacial characteristics of composites fabricated by laminated object manufacturing, Composites A 29A (1998) 1165–1174.

[24] A.G. Evans, M.Y. He, J.W. Hutchinson, Interface debonding and fiber cracking in brittle matrix composites, J. Am. Ceram. Soc. 72 (12) (1989) 2300–2303.

[25] M.-Y. He, J.W. Hutchinson, Kinking of a crack out of an interface, J. Appl. Mech. Trans. ASME 56 (1989) 270–278.

[26] M.-Y. He, A. Bartlett, A.G. Evans, Kinking of a crack out of an interface: role of in-plane stress, J. Am. Ceram. Soc. 74 (40) (1991) 767–771.

[27] A.G. Evans, D.B. Marshall, The mechanical behavior of ceramic matrix composites, Acta Metall. Mater. 37 (10) (1989) 2567–2584.

[28] A.G. Evans, M. Rühle, B.J. Dalgleish, P.G. Charalambides, The fracture energy of bimaterial interfaces, Mater. Sci. Eng. A A126 (1990) 53–64.

[29] M.I. Darby, Stress distribution near a crack crossing an interface, Int. J. Fract. 15 (4) (1979) 311–320.

[30] A.J. Phillipps, W.J. Clegg, T.W. Clyne, Fracture behavior of ceramic laminates in bending: II. Comparison of modeling with experimental data, Acta. Metall. Mater. 41 (3) (1993) 819–827.

[31] C.A. Folsom, F.W. Zok, F.F. Lange, Flexural properties of brittle multilayer materials: I. Modeling, J. Am. Ceram. Soc. 77 (3) (1994) 689–696.

[32] C.A. Folsom, F.W. Zok, F.F. Lange, Flexural properties of brittle multilayer materials: II. Experiments, J. Am. Ceram. Soc. 77 (8) (1994) 2081–2087.

[33] S.P. Yushanov, A.E. Bogdanovich, Analytical probabilistic modeling of initial failure and reliability of laminated composite structures, Int. J. Solids Struct. 35 (7/8) (1998) 665–685.

[34] T.Y. Kam, E.S. Chang, Reliability formulation for composite laminates subjected to first-ply failure, Compos. Struct. 38 (1–4) (1997) 447–452.

[35] D. Kovar, M. Thouless, Simple method for determining frictional sliding resistance and frictional energy dissipation in layered ceramics, J. Am. Ceram. Soc. 80 (3) (1997) 673–679.

[36] J.X. Zhang, D.L. Jiang, S.H. Tan, L.H. Gui, M.L. Ruan, Aqueous processing of SiC green sheets I. Dispersant, J. Mater. Res. 17 (11) (2002) 2012–2018.

[37] J.X. Zhang, D.L. Jiang, S.H. Tan, L.H. Gui, M.L. Ruan, Aqueous processing of SiC green sheets II. Binder and plasticizer, J. Mater. Res. 17 (11) (2002) 2018–2025.

[38] H.G. Tattersall, G. Tappin, The work of fracture and its measurement in metals ceramics and other materials, J. Mater. Sci. 1 (1966) 296–301.