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Effect of multilayer coating on mechanical properties of Nicalon-fibre-reinforced silicon carbide composites

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Nicalon-fibre-reinforced SiC composites were fabricated by combining polymer solution infiltration (PSI) and chemical vapour infiltration (CVI). Effect of multilayer coating on mechanical properties of the composites was investigated. The coatings consisted of chemically vapour deposited (CVD) C and SiC and were designed to enhance fibre pull-out in the composites. It was found that the flexural strength and fracture toughness of the composites were increased with the number of coating layers and was a maximum for 7 coating layers which consisted of C/SiC/C/SiC/C/SiC/C. Typical flexural strength and fracture toughness of the composites were 300 MPa and 14.5 MPa $m^{1/2}$, respectively.

1. Introduction

Fibre reinforcements in ceramics have been known to prevent catastrophic brittle failure by providing various energy dissipation processes during crack advance and improve the fracture resistance of ceramics [1,2]. Their ability to do so depends on fibre coatings that allow fibre pull-out through fibre debonding and frictional sliding in the matrix [3]. Thus, there is considerable interest in developing fibre coatings for ceramic matrix composites.

There are several possible strategies for enhancing fibre pull-out in ceramic matrix composites:

1. Weak interface coatings may be used. The weak interface may allow interface debonding and sliding and enhance fibre pull-out. Coatings of C and BN have been used to form a weak interface between fibre and matrix.
2. Porous oxide coatings may be used. The concept recognizes that porosity generally decreases the fracture energy of brittle materials. Consequently, introduction of porosity into a coating may satisfy debonding requirements and enhance fibre pull-out. Davis and co-workers [4] used porous oxide coatings on sapphire fibre. The porous oxide coatings satisfied the requirements for toughness improvement in sapphire fibre-reinforced alumina composites.
3. Multilayer coating can be used to enhance fibre pull-out. It consists of several thin layers that are weakly bonded to each other. The multilayer coating simulates the structure in carbon coating where the crystallographic layers are weakly bonded to one another in the thickness direction. This weak interface provides the low interfacial strength for debonding and sliding in the wake of crack advance. The weak bonding ensures that an advancing crack in the matrix will be deflected along one or more of the interfaces to promote fibre debonding, fricitional sliding, and eventual pull-out of fibres. One of the benefits of multilayer coating compared with the first two approaches is that the interfacial area to be debonded and slided is larger than those of the first two approaches.

Well known coatings that allow fibre debonding and frictional sliding are C and BN. Also, a SiC matrix composite layers may be attenuated with SiC layers and this eliminates any potential thermal stress problem which may influence the debonding and/or sliding characteristics. Clegg and co-workers [5] fabricated flexible sheets of SiC separated by thin carbon layers. In the material, the carbon layer serves as a weak interface, whereas the SiC phase is the load carrying phase. The major result of that work was the large increase in fracture toughness with retension of high strength. Therefore, we devized C/SiC multilayer coatings, in which the SiC was located outside, for the Nicalon-fibre-reinforced-SiC composites.

The purposes of this paper were to explore the applicability of C/SiC multilayer coating to ceramic matrix composites and to investigate the effect of multilayer coating on the mechanical properties of Nicalon-fibre-reinforced SiC composites.

2. Experimental procedure

Nicalon SiC yarn (approximately 500 filaments/yarn, filament diameter ~ 12 μm, Nippon Carbon Company, Tokyo, Japan) was cut into 6 cm lengths and put on a graphite substrate in a tube furnace. The fibres were then coated with C and SiC alternatively. CVD technique was used for fibre coating and processing

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parameters were determined from the earlier work [6]. The processing parameters for C coating were set at 1100 °C, a methane (CH_4) flow rate of 0.006 m^3h^{-1}, a hydrogen to methane ratio of 1, and a total pressure of 100 kPa. The processing parameters for SiC coating were set at 1150 °C, a methyltrichlorosilane (CH_3SiCl_3, MTS) flow rate of 0.006 m^3h^{-1}, a hydrogen to MTS ratio of 10, and a total pressure of 5 kPa.

Nicalon fibre coated with 1(C), 2(C/SiC), 3(C/SiC/C), 7(C/SiC/C/SiC/C/SiC/C), and 11 (C/SiC/C/SiC/C/SiC/C/SiC/C/SiC/C) layers were prepared. After coating, the coated layers were observed and analysed using a scanning electron microscopy (SEM) and X-ray diffraction (XRD). The multilayer coated Nicalon fibre was tied together in a bundle shape. The preforms were infiltrated via PSI with a boiling polycarbosilane solution (70 wt % solution in hexane) for 20 min. The infiltrated preforms were then heated to 600 °C in flowing argon to render the polycarbosilane insoluble. After the 6 PSI/heat-treatment cycles, the preforms were pyrolysed completely at 1000 °C under flowing argon.

The preforms were then cut into 3 × 4 × 40 mm bars and their surfaces and edges were polished with a 800-grit diamond wheel. The bars were then infiltrated by CVI with methyltrichlorosilane (CH_3SiCl_3, MTS) and hydrogen to form CVD SiC. The processing parameters for CVI were identical with the conditions for SiC fibre coating. Details of PSI/CVI process are given in earlier works [2].

Three point flexural strength and fracture toughness were measured using plain bars and single-edge-notched bars whose notch (0.3 mm wide and 1.5 mm deep) was made in the plane normal to the fibre orientation.

3. Results and discussion

3.1. Characterization of coating

The microstructures of the coated fibre are shown in Fig. 1. Although the outer surface of the SiC coating has a rough appearance, the carbon coating reduces the roughness of the SiC coating layer. It also shows that the fibre was uniformly coated with the 7 layers (C/SiC/C/SiC/C/SiC/C).The thickness of each coating layer was 0.05-0.1 µm for C and 0.4-0.5 µm for SiC. XRD of the coating layers showed crystalline graphite for C coating and β-SiC for SiC coating.

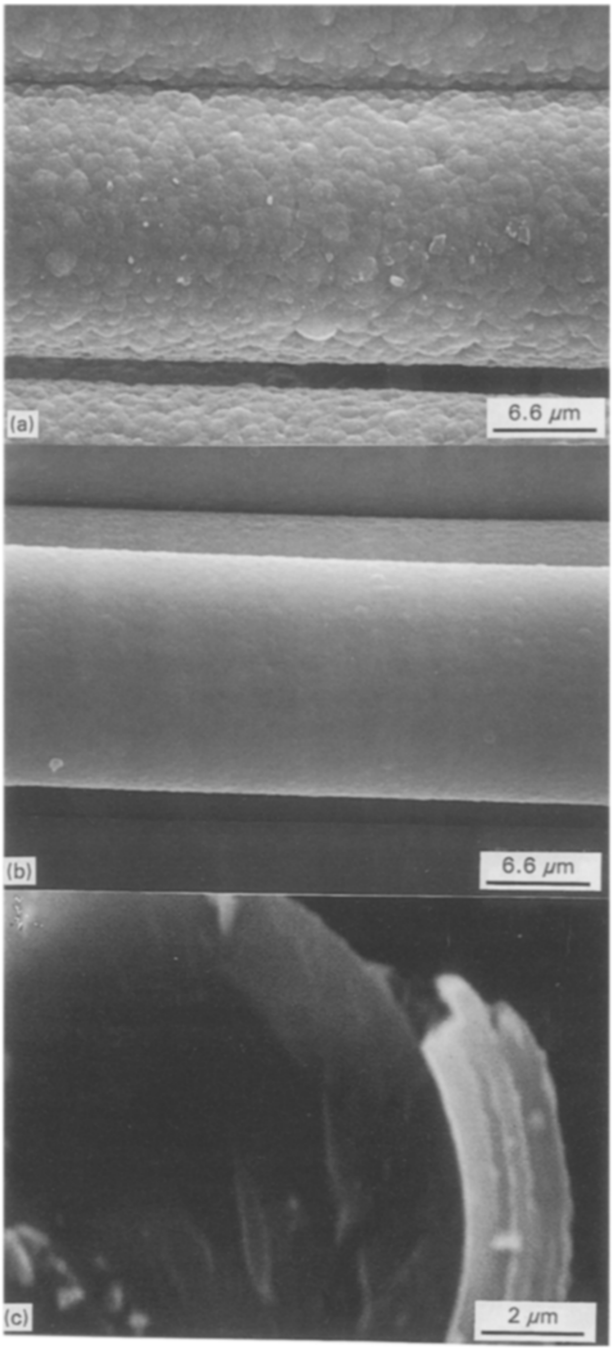


Figure 1 Scanning electron micrographs of multilayer coated Nicalon fibre showing (a) C/SiC surface, (b) C/SiC/C surface, and (c) cross-section of the 7 layer coatings.

3.2. Fracture morphology

Fig. 2 shows the SEM of a pulled-out 7 layer coated fibre after flexure. As shown, there are many fragments of coating layers, which indicate the occurrence of active crack-multi-interface interaction and the excellent failure mode in ceramic matrix composites. The advancing crack in the matrix was deflected along multi-interfaces, and the interfaces debonded, and many segments of coating layers broke away from the fibre. It proves that the individual interfaces act as debonding paths and slide in the wake of the crack. It also proves that multilayer coating is applicable to the fibre-reinforced-ceramic matrix composites.

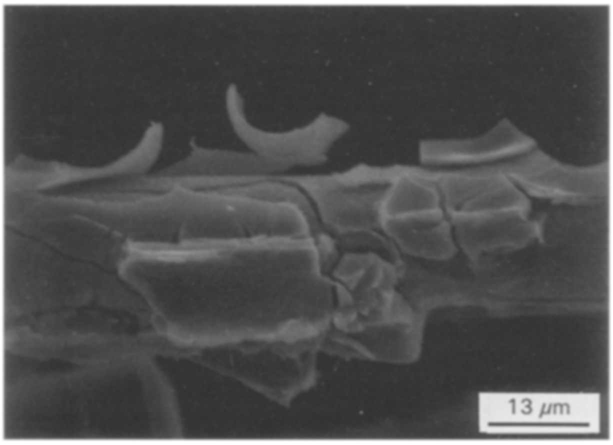
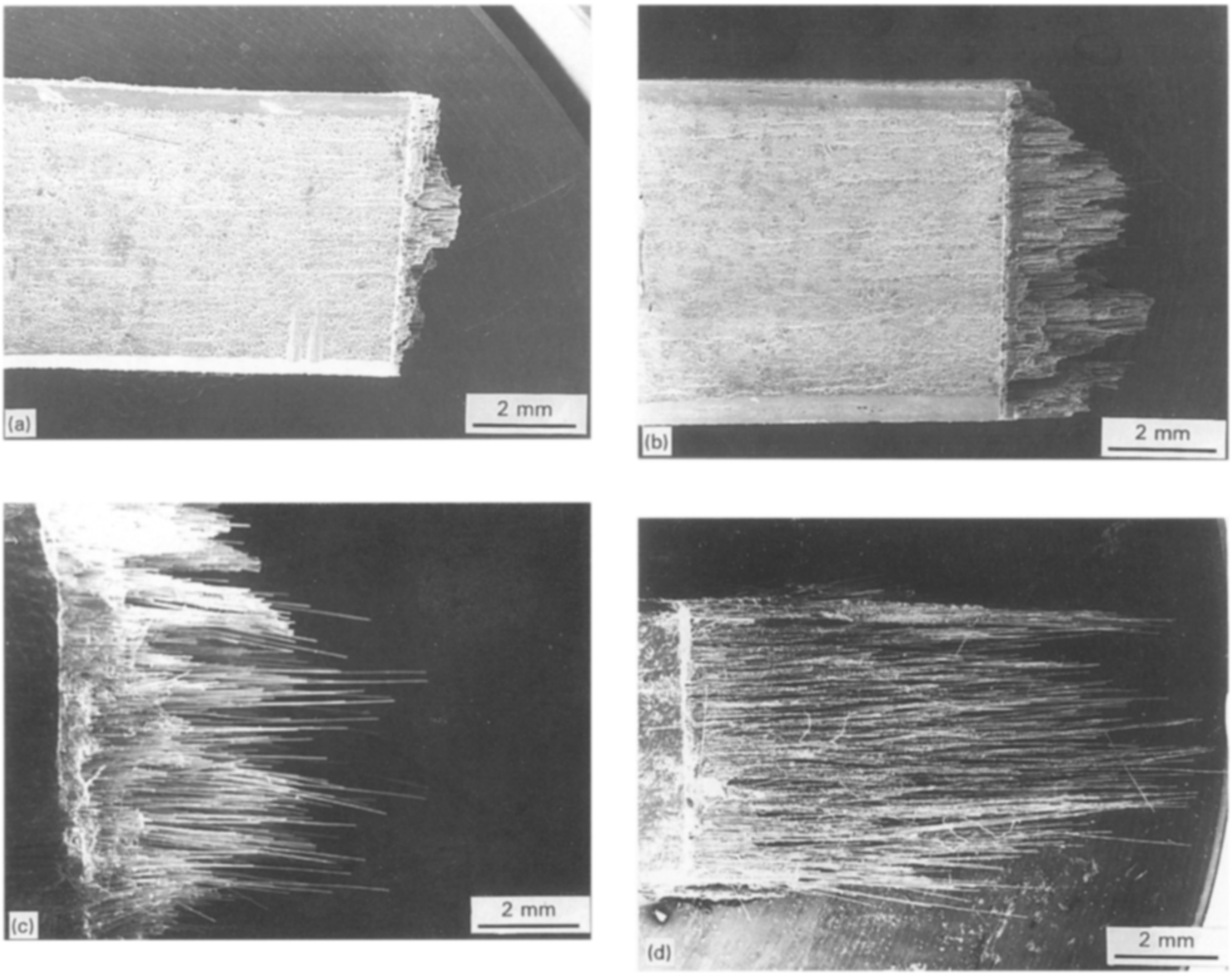


Figure 2 Scanning electron micrograph of the pulled-out 7 layer coated fibre after flexure.



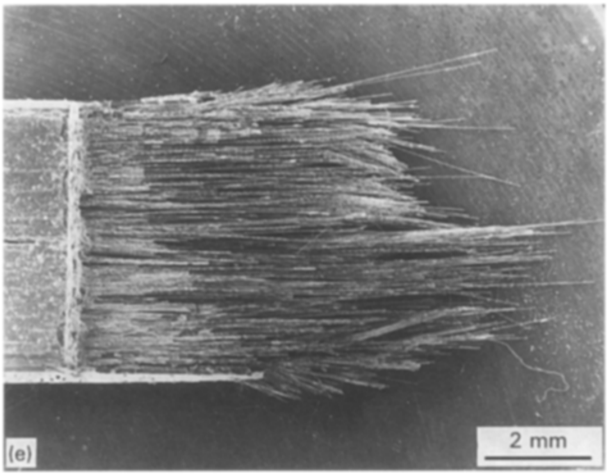


Figure 3 Scanning electron micrographs of the fracture surfaces of Nicalon-fibre-reinforced SiC composites with various fibres: (a) uncoated, (b) 1 layer coated, (c) 3 layer coated, (d) 7 layer coated, and (e) 11 layer coated.

For observing the pull-out length of the fibres, the fracture surfaces of Nicalon-fibre-reinforced SiC composites with various fibres were observed by SEM (Fig. 3). As expected, the length of fibre pull-out was increased with the number of fibre coatings. It can be explained that the advancing crack firstly meets the outer coating layer, debonds the interface, deflects along the interface, and when it meets a defect in the SiC coating layer during propagation, it penetrates the layer and meets another interface and propagates in the same manner. Hence, the multilayer coating increases the pull-out length of fibres and the increased pull-out may lead to the improved mechanical properties.

3.3. Mechanical properties

The mechanical properties of Nicalon-fibre-reinforced SiC composites fabricated using 0, 1, 3, 7, and 11 layer coated fibres are summarized in Table I. As expected, it was found that flexural strength and fracture toughness increased with the number of coating layers, and reached a maximum at 7 layers. The improvement of mechanical properties with the number of coating layers was due to the increased crack–interface interaction and increased pull-out of fibres, as shown in Figs 2 and 3. It is further supported by typical load–displacement curves for Nicalon-fibre-reinforced SiC composites with various fibres which is shown in Fig. 4. It shows typical delayed fracture behaviour of fibre-reinforced composites. However, the composites with 11 coating layers showed lower flexural strength and fracture toughness than the composites with 7 coating layers. It may be due to the presence of too many weak interface areas in the composites. Hence, there is an optimum number of coating layers in the fabrication of composites using multilayer coating. It may depend on both the thickness and the bond strength of each coating layer.

Typical flexural strength of 300 MPa was obtained in Nicalon-fibre-reinforced SiC composites with

TABLE I Mechanical properties of Nicalon-fibre-reinforced SiC composites with various fibres

Number of coating layer	Flexural strength[a] (MPa)	Fracture toughness[a] (MPa $m^{1/2}$)
0	73 ± 10	3.6 ± 0.6
1	262 ± 30	8.9 ± 0.7
3	274 ± 58	11.4 ± 0.8
7	302 ± 42	14.5 ± 1.0
11	282 ± 23	10.8 ± 0.9

[a] Average of 6 samples.

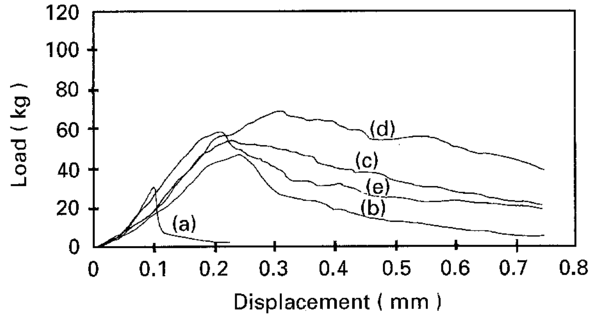


Figure 4 Load–displacement curves for the Nicalon-fibre-reinforced SiC composites with various fibres: (a) uncoated, (b) 1 layer coated, (c) 3 layer coated, (d) 7 layer coated, and (e) 11 layer coated.

7 coating layers. This value is comparable or slightly lower than that of monolithic SiC which has typical values of 300–500 MPa [7,8]. However, a fracture toughness of 14.5 MPa $m^{1/2}$ was obtained in the composites and it demonstrates a quadrupling of the fracture toughness over monolithic SiC which has typical values of 2.5–4 MPa $m^{1/2}$ [9,10].

4. Conclusions

1. The C/SiC multilayer coating is applicable to fibre-reinforced-ceramic matrix composites for providing excellent fibre pull-out.

2. Flexural strength and fracture toughness of Nicalon-fibre-reinforced SiC composites with multilayer coated fibre were increased with the number of coating layers due to the increased crack–interface interaction and the increased pull-out of fibres.

3. Typical flexural strength and fracture toughness of Nicalon-fibre-reinforced SiC composites, which was fabricated via PSI/CVI using 7 layer (C/SiC/C/SiC/C/SiC/C) coated fibres, were 300 MPa and 14.5 MPa $m^{1/2}$, respectively.

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