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Nicalon-fibre-reinforced silicon-carbide composites via polymer solution infiltration and chemical vapour infiltration

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A new, faster process was developed for the fabrication of Nicalon-fibre-reinforced SiC composites by combining polymer solution infiltration (PSI) and chemical vapour infiltration (CVI). The process led to the near-net-shape fabrication of fibre-reinforced ceramic-matrix composites and reduced infiltration time. Typical flexural strength and fracture toughness of these composites were 296 MPa and 10.9 MPa $m^{1/2}$ at room temperature (RT) and 252 MPa and 9.6 MPa $m^{1/2}$ at 1000 °C, respectively. The composites exhibited load-carrying capability after crack initiation.

1. Introduction

Fibre-reinforced-ceramic-matrix composites have recently attracted attention for use in high-temperature structural applications [1, 2]. The primary reason for this interest lies in the assumption that strong ceramic fibres can prevent catastropic brittle failure in ceramics by providing various energy-dissipation processes during crack advance [3]. Early works utilizing ceramic fibres in ceramic matrices demonstrated the potential of this approach [4–10].

However, a generic problem that must be overcome is that some ceramic fabrication processes tend to mechanically and chemically damage the fibres when they are consolidated within a ceramic matrix. For example, the fibres may be broken by a pressing operation, or the high sintering temperature required to densify the ceramic matrix may damage the fibres or cause them to react chemically with the matrix. The most common processing method for overcoming the above problems is CVI. Although this method does not damage the fibres during composite fabrication, it requires an excessively long infiltration time (100–300 h). Recently, a new process for reducing the infiltration time to 12–30 h was developed by combining a thermal gradient and a forced-flow approach [6–8]. However, this process limits the shapes of fibre preform that could be used.

The objective of this work was to fabricate Nicalon-fibre-reinforced SiC composites by infiltrating the fibre preform using polycarbosilane in liquid solution (a form of PSI) and subsequently infiltrating the composite using methyltrichlorosilane (CH_3SiCl_3, MTS) in gaseous phase (i.e. CVI). This new process (PSI/CVI) can lead to near-net-shape fabrication of fibre-reinforced-ceramic-matrix composites and significantly reduce the infiltration time.

2. Experimental procedures

Nicalon SiC yarn* heat treated at 500 °C for 20 min in an argon atmosphere was cut and tied together in a bundle shape. The fibres in the preform were then precoated with a thin layer of carbon (~1 μm) to weaken the bonds between the fibres and the matrix to enhance the desired fibre pull-out. The carbon was deposited by infiltrating the preforms with 0.1 MPa methane at 1100 °C for 1 h.

The carbon-coated 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 desired PSI/heat-treatment cycles, the preforms were pyrolysed completely at 1000 °C under flowing argon.

The preforms were then cut into 4 × 5 × 50 mm bars and their surfaces and edges were polished with an 800-grit diamond wheel for flexural testing. Half of the bars were then infiltrated by CVI with MTS and hydrogen to form chemical-vapour-deposited SiC. Based on earlier work [11], the processing parameters for CVI were set at 1150 °C, a MTS flow rate of 100 s.c.c.m, a hydrogen-to-MTS ratio of 10, and a total pressure of 5 kPa.

Densities of composites were measured using the Archimedes principle, and the fibre volume fraction was determined by a linear-intercept method. Three-point flexural strength and fracture toughness were measured at RT and 1000 °C using plain bars and single-edge-notched beams 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. Density increase by infiltration

Fibre contents in the preforms were 57–62 vol %. The

* Approximately 500 filaments/yarn, filament diameter ~ 12 μm, Nippon Carbon Company, Tokyo, Japan.

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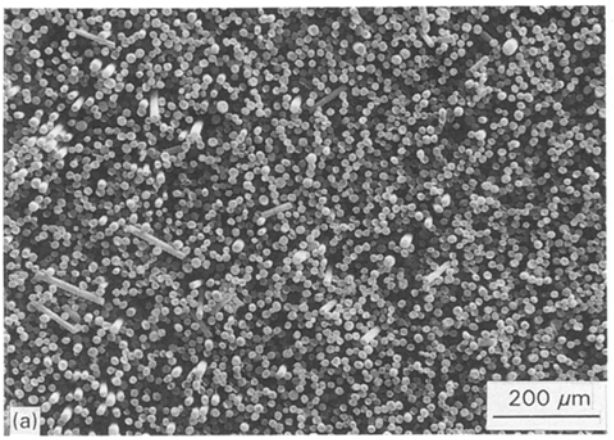

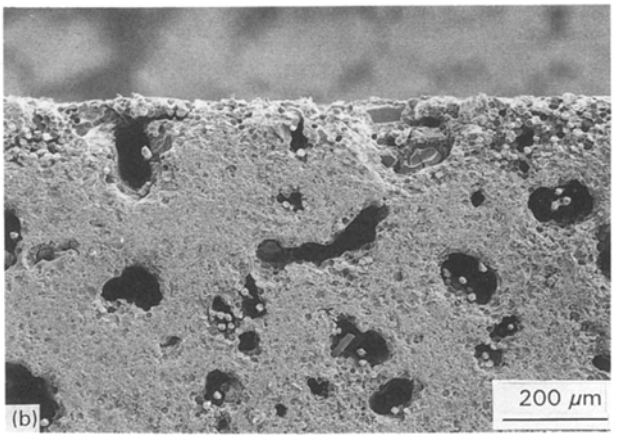

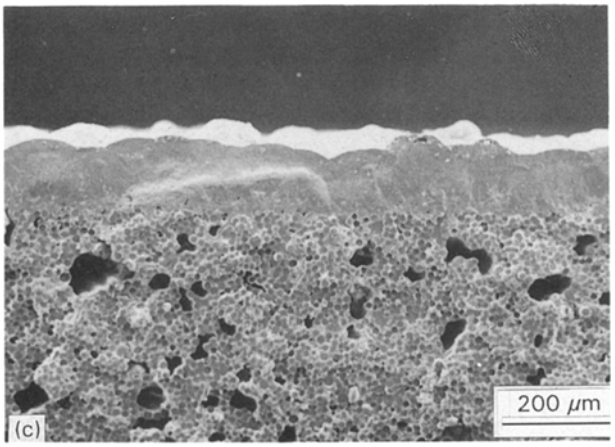

Figure 1 The cross-sections of composites after each process step: (a) as-prepared Nicalon preform after carbon coating, (b) after six cycles of PSI, and (c) after six cycles of PSI and 12 h of CVI.

densities of the composites increased with the number of PSI cycles. A density of 2000 kg m^{-3} (71% TD) was obtained after six cycles. Further cycles were not recommended due to the rapidly decreasing effectiveness of infiltration as reported in earlier work [12].

The densities of composites after PSI further increased up to 2310 kg cm^{-3} (82% TD) after CVI. Density increases by matrix build-up via PSI and CVI are clearly shown in Fig. 1, which shows the cross-sections of composites after each process step.

3.2. Room temperature mechanical properties

The mechanical properties of PSI and PSI/CVI composites are summarized in Table I. The composites show significant strength development even after PSI. Observation by scanning electron microscopy (SEM) and X-ray diffraction (XRD) indicates that the matrix built-up from pyrolysis of polycarbosilane consists of sintered β-SiC form. Flexural strength values after PSI/CVI are lower than those of monolithic SiC which has typical values of 300–500 MPa [13, 14] due to the presence of large pores in the range of 100–150 μm as shown in Fig. 1, which were observed as fracture origins. A fracture toughness of 10.6 MPa m$^{1/2}$ was obtained in PSI/CVI composites. This value demonstrates a tripling of the fracture toughness over monolithic SiC which has typical values of 3–4 MPa m$^{1/2}$ [13]. This improvement of toughness was due to the pull-out of fibres, as shown in Fig. 2.

The pull-out mechanism is further supported by a typical load–displacement curve for PSI/CVI composites which is shown in Fig. 3. It shows typical delayed fracture behaviour of fibre-reinforced composites which involves matrix fracture followed by strength due to the support of the fibres.

3.3. Elevated-temperature mechanical properties

As expected, samples treated via PSI only showed considerable loss of mechanical properties at 1000 °C

TABLE I Properties of Nicalon-fibre/SiC composites at each process step

Sample	Density (kg m^{-3})	Flexural strength[a] (MPa)		Fracture toughness[a] (MPa m$^{1/2}$)	
		RT	1000 °C	RT	1000 °C
As-prepared Nicalon preform after carbon coating	1530 (60% TD)	–	–	–	–
Nicalon/SiC composites after 6 cycles of PSI	2000 (71% TD)	224 ± 27	134 ± 38	6.2 ± 0.5	4.1 ± 0.4
Nicalon/SiC composites after 6 cycles of PSI and 12 h of CVI	2310 (82% TD)	296 ± 35	252 ± 31	10.9 ± 0.7	9.6 ± 0.9

[a] Average of five samples.

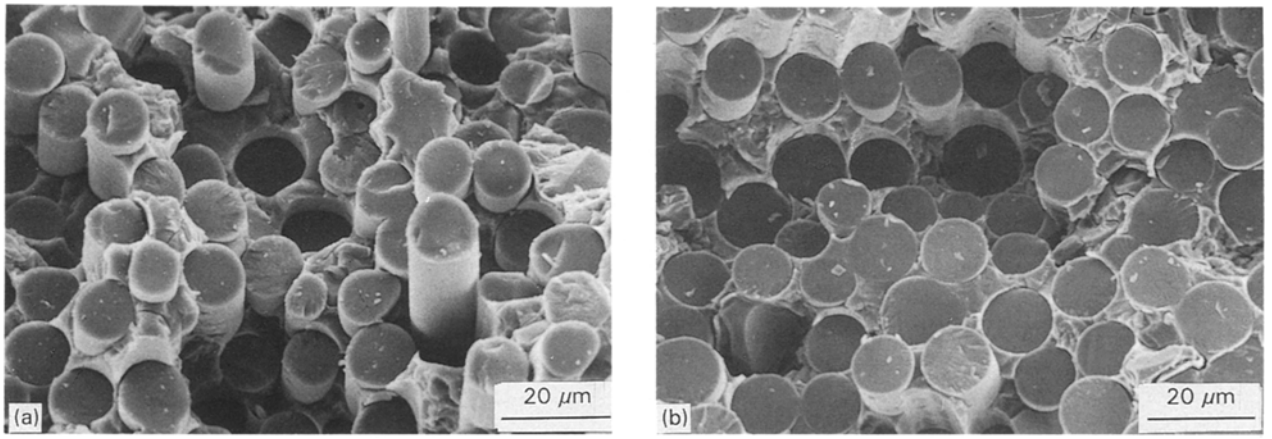

Figure 2 Scanning electron micrographs of: (a) the RT and (b) 1000 °C fracture surface of Nicalon-fibre/SiC composites via PSI/CVI.

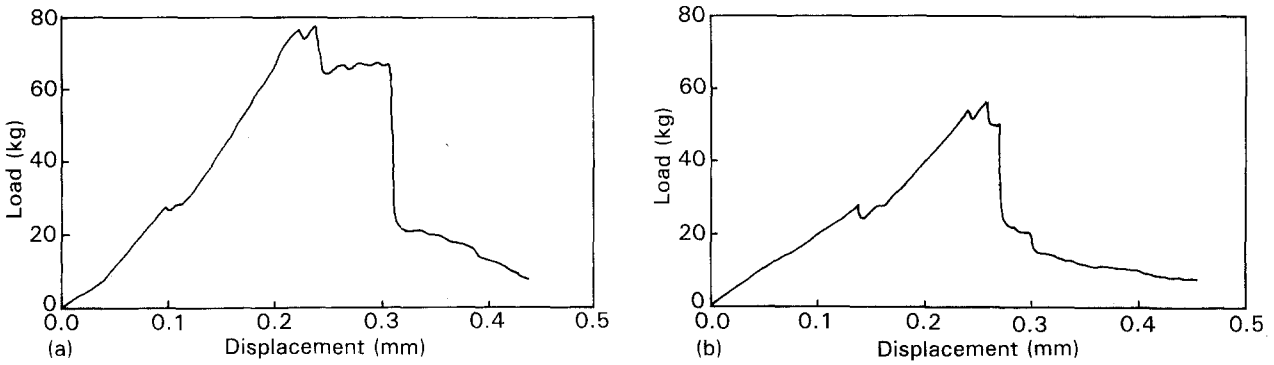

Figure 3 Load versus displacement curves at: (a) room temperature and (b) 1000 °C for the Nicalon-fibre/SiC composites via PSI/CVI.

due to their high porosities which make the composites vulnerable to oxidation. However, PSI/CVI composites showed little change in mechanical properties due to the lower porosities and the presence of a protective surface layer formed by CVI, as shown in Fig. 1. This means that the CVI step makes the composites effectively impermeable. A slight loss of the mechanical properties of PSI/CVI composites at elevated temperature is likely due to the degradation of fibre strength. Early work on the strength of Nicalon fibres at elevated temperatures indicates that degradation of fibres may well occur [15–17].

4. Conclusions

A new, faster, near-net-shape PSI/CVI fabrication process was demonstrated for the fabrication of SiC-fibre–SiC-matrix composites. Composites so prepared had an average density of 82% TD. Typical flexural strength and fracture toughness were 296 MPa and 10.9 MPa $m^{1/2}$ at RT and 252 MPa and 9.6 MPa $m^{1/2}$ at 1000 °C, respectively. And the composites exhibited load-carrying capability after crack initiation and toughening by a fibre-pull-out mechanism.

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