

# Continuous fibre reinforced mullite matrix composites by sol–gel processing

## Part II *Properties and fracture behaviour*

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The mechanical and interfacial properties of the mullite matrix and alumina–silica matrix composites reported in a previous publication [1], are related to the processing parameters. The flexural strengths were in the range of 428–737 MPa, flexural moduli from 82–214 GPa, work of fracture values from 293–482 KJ m<sup>-2</sup> and ultimate failure strains from 0.16–0.45%. The interfacial strengths were determined by the fibre push-in test and compared with fracture behaviour. The formation of different interfaces in the composites and the influence of the density of thermally-induced cracks and porosity on flexural modulus is discussed.

### 1. Introduction

The toughening effect of fibres in a ceramic matrix composite depends both on the nature of the interface between fibres and matrix and on the retained strength of the fibres after consolidation [2–4]. The nature of the interface is governed by the processing conditions as well as the starting materials for the matrix and the type of reinforcing fibres. These factors can significantly affect the interfacial strength and may lead to the formation of the preferred state of a relatively weak interface [5, 6], and in some cases pores or crevices along the interfaces can also result in a low interfacial strength [7]. In this paper, the properties and fracture behaviour of the composites described in Part I of this work [1] are examined with respect to processing conditions. Interfacial strengths between the fibres and matrix within the composites were determined from the fibre push-in test and compared to their fracture behaviour.

### 2. Experimental procedures

The fabrication of the composites was described in Part I of this work [1]. The process conditions are summarized in Table I. The spacing of matrix cracks in polished longitudinal sections of the fractured composite samples was determined by computer-controlled image analysis using an Ams Optomax V. The fibre push-in test was performed with an automatic unloading type Leco M-400 Hardness Tester with a diamond indenter. A loading range of 0.1–5 N was used with a load duration of 15 s. For the test the composite samples were mounted in a thermosetting resin, and were polished to a thickness of 10 mm. The flexural strengths, flexural moduli, and work of fracture of the composites at room temperature were determined using a three point bending test on a Mayes Universal Tester at a displacement rate of

0.2 mm min<sup>-1</sup>. The span of the lower support pins was 30 or 40 mm, and a span to specimen thickness ratio greater than 20 was employed. Rectangular test specimens, cut from hot-pressed cylindrical composite samples, were ground and polished to the dimensions of 37 or 47 mm in length, 2.0 mm in width and 1.5 mm in thickness. The load–deflection curves for the composites were recorded to fracture. The mechanism of failure was identified by visual examination after the test and by scanning electron microscope (SEM) examination of the fracture surface of tested specimens.

During the test, most of the composites initially showed an elastic response with the deflection increasing linearly with an increase in the load, followed by an extended non-elastic regime. Generally after the maximum value of the load was reached, the degree of subsequent extension was strongly dependent on the nature of the interface between fibre and matrix.

The flexural strengths and moduli were determined according to British standard methods of testing [8]. The work of fracture was calculated from the area under the load–deflection curve. The ultimate failure strain,  $\epsilon$ , was calculated by the approximate Equation 1 where  $l$  is the distance between the lower supporting points and  $D$  is the deflection at fracture.

$$\epsilon = \frac{(l^2 + 4D^2)^{1/2} - l}{l} \quad (1)$$

### 3. Results

#### 3.1. Flexural strength, modulus, ultimate failure strain and work of fracture

Fig. 1(a–d) are representative examples of the stress versus deflection curves for the composites, compared with typical results for the pure matrix materials (dotted lines). A summary of hot-pressing parameters together with the properties of the composites is given

TABLE I Summary of the hot-pressing conditions and properties of composites

Matrix starting material	$\alpha$ -alumina/ Ludox silica	$\alpha$ -alumina/ Ludox silica	$\alpha$ -alumina/ Ludox silica	$\alpha$ -alumina/ Ludox silica	$\alpha$ -alumina/ Ludox silica	Boehmite/ Ludox silica
Hot-pressing conditions	1300 °C 1 h 25 MPa	1550 °C 0.5 h 15 MPa	1550 °C 0.5 h 15 MPa	1550 °C 1.5 h 15 MPa	1400 °C 0.5 h 18 MPa	1300 °C 40 min 15 MPa
Fibre/matrix	C/(A + S)	C/M1	SiC/M1a	SiC/M1b	SiC/(A + S)	C/M2
Number tested	12	6	6	6	–	8
$V_f$ ( $\pm 2\%$ )	40	52	60	32	32	48
$\rho_c/\rho_{c0}$ (%)	98	95	98	99	98	84
$\sigma$ (MPa)	720 $\pm$ 156	737 $\pm$ 128	630 $\pm$ 98	203 $\pm$ 74	50	428 $\pm$ 79
$E$ (GPa)	107 $\pm$ 33	127 $\pm$ 64	208 $\pm$ 11	214 $\pm$ 103	–	82 $\pm$ 39
$W$ (kJ m $^{-2}$ )	318 $\pm$ 69	482 $\pm$ 122	293 $\pm$ 111	43 $\pm$ 25	–	350 $\pm$ 90
$\varepsilon$ (%)	0.16 $\pm$ 0.03	0.37 $\pm$ 0.13	0.30 $\pm$ 0.10	0.06 $\pm$ 0.02	0	0.45 $\pm$ 0.14
$\sigma_c$ (GPa)	1.7	2.2	1.6	0.9	–	2.0
$E_c$ (GPa)	120	160	149	122	–	157
$\alpha$ (see text)	0.33	0.37	0.32	–	–	0.44
Fracture mode	Non-brittle	Non-brittle	Non-brittle	Brittle	Brittle	Non-brittle
Interfacial nature	Weak	Weak	Weak	Strong	Strong	Weak

(A + S): a matrix composed of  $\alpha$ -alumina particles dispersed in a silica glass phase with a  $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$  composition; M1: a mullite matrix produced from  $\alpha$ -alumina powder and Ludox silica colloid; M2: a mullite matrix produced from boehmite gel powder and Ludox silica colloid;  $\pm$ : standard deviation.

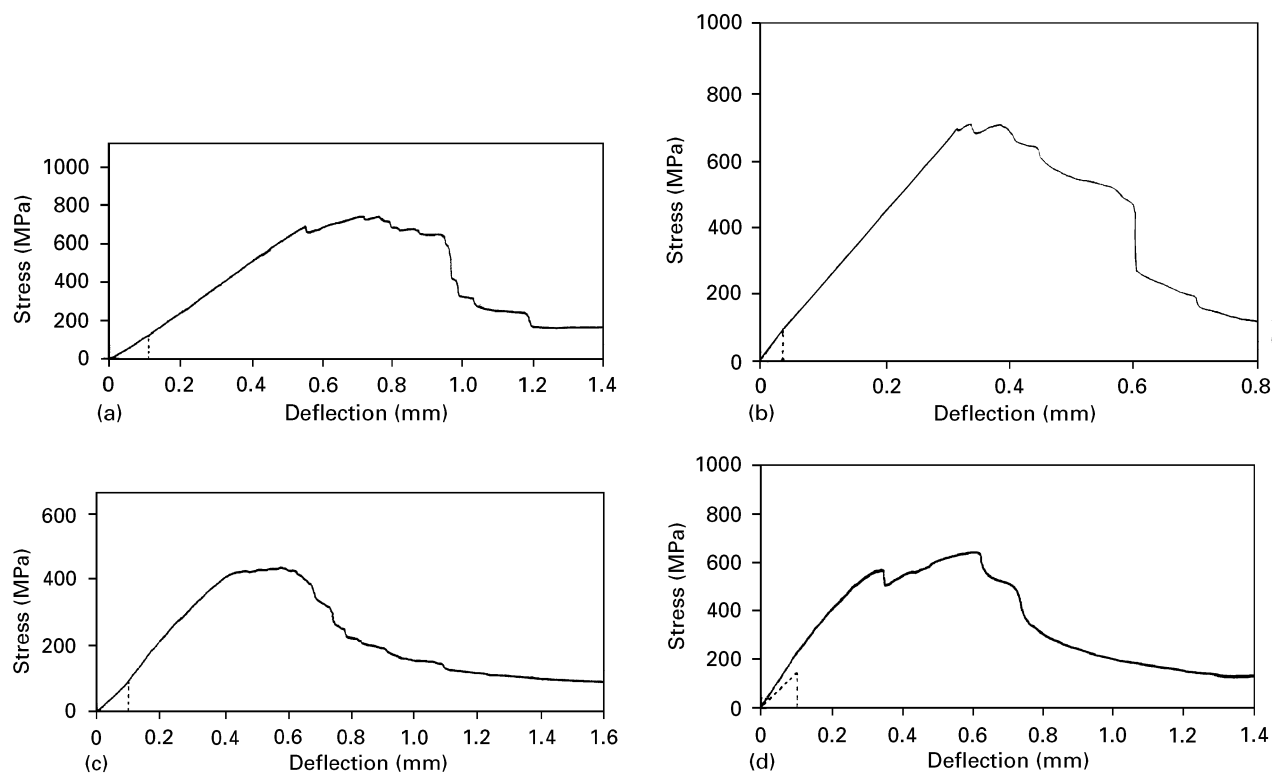


Figure 1 Typical load versus deflection plots for composites tested by three-point-bending; behaviour of matrix shown by dotted curves. (a) Carbon fibre mullite matrix composite C/M1 hot-pressed at 1550 °C for 0.5 h at 15 MPa, (b) carbon fibre alumina-silica mixture matrix composite C/(A + S) hot-pressed at 1300 °C for 1 h at 25 MPa, (c) carbon fibre mullite matrix composite C/M2 hot-pressed at 1300 °C for 40 min at 15 MPa from boehmite and colloidal silica; and (d) Nicalon-SiC fibre mullite matrix composites SiC/M1a hot-pressed at 1550 °C for 0.5 h at 15 MPa.

in Table I.  $V_f$  is the measured fibre volume fraction,  $\rho_c/\rho_{c0}$  the ratio of measured to theoretical density expressed as a percentage,  $\sigma$  the ultimate flexural strength,  $E$  the flexural modulus,  $W$  the work of fracture, and  $\varepsilon$  the ultimate failure strain.  $\sigma_c$  and  $E_c$  are the ultimate tensile strength and modulus of the composites calculated theoretically. According to theory [9], above a critical fibre volume fraction the ultimate tensile strength of a brittle matrix composite ( $\sigma_c$ ) con-

taining continuous fibre with a unique strength, is given by:

$$\sigma_c = V_f \sigma_{fu} \quad (2)$$

where  $\sigma_{fu}$  is the ultimate tensile strength of fibres. The initial tensile modulus will depend upon the load bearing capability of the matrix, which will be a function of the number of the thermally induced matrix cracks, and the ability of the interface to transfer stress

between the components. The lower bound value is given by:

$$E_c = V_f E_f \quad (3)$$

whereas the higher bound value is given by

$$E_c = V_f E_f + E_m V_m \quad (4)$$

Here  $E_f$  and  $E_m$  are the elastic moduli of the fibre and matrix respectively, and  $V_m$  the matrix volume fraction. The  $\sigma_c$  and  $E_c$  values listed in Table I were calculated from Equations 2 and 3 respectively, using the fibre properties given in Part I of this work [1].

Thus the strength of a fibre composite is a complex function of the statistical properties of the fibre and matrix. Furthermore the efficiency of the fibre–matrix interface to transfer the additional stress thrown onto the fibre when the matrix cracks determines whether brittle or ductile behaviour is observed. Interfacial debonding adjacent to a matrix crack diffuses the additional load over a large volume, leading to tougher behaviour. With a strong interface, the stress concentrating effect of a matrix crack can lead to the fracture of the adjacent fibre and the propagation of one crack, leading to brittle failure. In the flexural test employed here, only the outside surface experiences a maximum stress. All failures were initiated on the tensile face of the specimen. Hannant [10] and Aveston *et al.* [11] have discussed the relationship between flexural strength and ultimate tensile strength of brittle matrix continuous fibre composites. In this study, the parameter  $\alpha (= E_m V_m / E_f V_f)$  [11], is 0.32 to 0.44, see Table I indicating according to Aveston *et al.* [11] that the ratio of the flexural strength to ultimate tensile strength of the composites is about 1.0 so the two strengths can be directly compared.

Referring to Table I, the highest average flexural strength ( $737 \pm 128$  MPa) was observed for the carbon fibre reinforced mullite matrix composite hot-pressed at  $1550^\circ\text{C}$  for 0.5 h at 15 MPa (C/M1). This composite also exhibited the highest work of fracture,  $482 \pm 122$   $\text{kJ m}^{-2}$ , and ultimate failure strain of  $0.37 \pm 0.13\%$ . These values were considerably higher than those for unreinforced bulk mullite of 150 MPa for  $\sigma$ ,  $5.18$   $\text{kJ m}^{-2}$  for the work of fracture and 0.001% for  $\epsilon$  [13]. Samples of bulk mullite were prepared by pressureless sintering of gels of  $\alpha$ -alumina and Ludox silica at  $1600^\circ\text{C}$  for 2 h to produce a density of  $3.1 \times 10^3$   $\text{kg m}^{-3}$  which is approximately 98% of the theoretical density for mullite of  $3.17 \times 10^3$   $\text{kg m}^{-3}$  [12, 13]. The carbon fibre reinforced composite with a matrix composed of  $\alpha$ -alumina particles dispersed in silica glass, C/(A + S) hot-pressed at the lower temperature of  $1300^\circ\text{C}$  so that no mullite was produced, had similar properties to the C/M1 composite hot-pressed at  $1550^\circ\text{C}$ . The SiC fibre mullite matrix composite (SiC/M1a) hot-pressed at  $1550^\circ\text{C}$  for 0.5 h also had a high strength ( $\sigma$ ) of  $630 \pm 98$  MPa, a  $W$  of  $293 \pm 111$   $\text{kJ m}^{-2}$ , and a value  $\epsilon$  of  $0.30 \pm 0.1\%$ . However, increasing the hot-pressing time to 1.5 h at  $1550^\circ\text{C}$  led to marked deterioration in the strength to  $203 \pm 74$  MPa, in  $W$  to values of  $43 \pm 25$   $\text{kJ m}^{-2}$ , and also in  $\epsilon$  to values of  $0.06 \pm 0.02\%$ . Also a change from non brittle fracture behaviour to brittle was

observed although the fibre volume fraction was lower than in the former case. Finally the carbon fibre mullite matrix composite prepared from boehmite and Ludox by method B in Part I of this work [1] and hot-pressed at only  $1300^\circ\text{C}$ , in spite of its relatively low overall densification (only 84%) still retained relatively high values for the strength of  $428 \pm 79$  MPa, for  $W$  of  $350 \pm 90$   $\text{kJ m}^{-2}$  and for  $\epsilon$  of  $0.45 \pm 0.14\%$ .

### 3.2. Observations of fracture surfaces

The nature of the fibre/matrix interface can be revealed by examining the fracture surfaces of composite specimens after the three point test. SEM micrographs are shown in Fig. 2(a–e). Extensive fibre pull-out was observed in the fracture surfaces of the carbon fibre composites, C/M1 and C/(A + S) with C/M1 exhibiting the greatest degree of fibre pull-out (Fig. 2a).

In the case of the SiC fibre composites, the SiC fibre/mullite composite, SiC/M1a, hot-pressed for 0.5 h showed non-brittle fracture behaviour with extensive fibre pull-out in the fracture surface (Fig. 2c). However, after hot-pressing for 1.5 h the SiC/M1b composite exhibited brittle fracture with only limited evidence of fibre pull out (Fig. 2d). The fracture surface for a SiC/(A + S) specimen is shown in Fig. 2e. This material was hot-pressed at  $1400^\circ\text{C}$ , for 0.5 h at 18 MPa. Poor strength was observed because of strong interfacial bonding, as discussed in Part I of this work [1]. No detailed property measurements were carried out for this material. These results indicate that the three SiC composites had significantly different interfacial states.

### 3.3. Interface characterization

The different states of bonding between the fibres and matrices for the three SiC fibre composites were also indicated by SEM observations of the indentations produced by the fibre push-in test. Fig. 3(a and b) refer to the SiC/M1a and SiC/M1b composites hot-pressed for 0.5 h and 1.5 h respectively and Fig. 3c to the SiC/(A + S) composite hot-pressed at  $1400^\circ\text{C}$ . The tests on the two SiC/M1 composite specimens were carried out under a load of 2 N, whereas for the SiC/(A + S) composite a load of 5 N was used. The SiC fibres in the mullite matrix were clearly pushed into the matrix surface as shown by Fig. 3(a and b). The results show that the interfacial bonding is lower between the fibre and the matrix in the SiC/M1a composite hot-pressed for 0.5 h compared with the composite hot-pressed for 1.5 h because a deeper indentation was observed in the former specimen (Fig. 3a). This was confirmed by repeated observations. However, in the case of the SiC/(A + S) composite, even under a load of 5 N, the fibre section was not pushed into the A + S matrix. Under this load, the fibre was fractured, leading to radical cracking of the adjacent matrix, as a result of the strong interfacial bond in this case (Fig. 3c).

The interfacial bond strength between the fibre and the matrix of the SiC composites was calculated from the indentations on the fibre and matrix and the

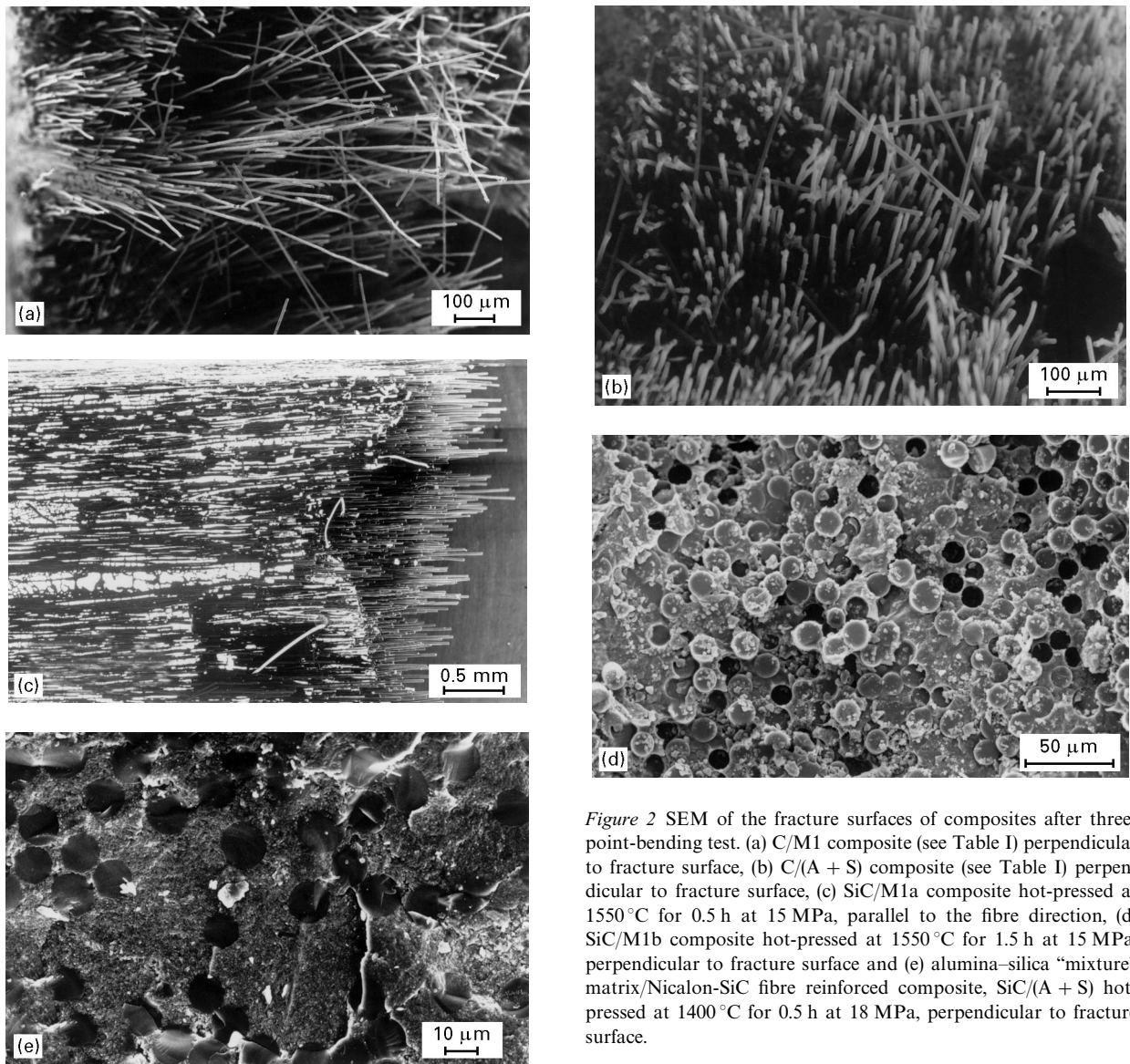


Figure 2 SEM of the fracture surfaces of composites after three-point-bending test. (a) C/M1 composite (see Table I) perpendicular to fracture surface, (b) C/(A + S) composite (see Table I) perpendicular to fracture surface, (c) SiC/M1a composite hot-pressed at 1550 °C for 0.5 h at 15 MPa, parallel to the fibre direction, (d) SiC/M1b composite hot-pressed at 1550 °C for 1.5 h at 15 MPa, perpendicular to fracture surface and (e) alumina-silica “mixture” matrix/Nicalon-SiC fibre reinforced composite, SiC/(A + S) hot-pressed at 1400 °C for 0.5 h at 18 MPa, perpendicular to fracture surface.

applied force according to the method of Marshall [14]. The interfacial bond strength values,  $\tau$ , are given in Table II for the SiC/M1 composites.

### 3.4. Multiple matrix cracking

Examining the polished surfaces of the carbon fibre composite specimens, C/(A + S) and C/M1, before and after a three point bend test, revealed that there was no change in the number of matrix cracks. This indicated that the matrix cracking, as a result of a mismatch between the thermal expansion coefficients of fibre and matrix had “saturated” on cooling from the processing temperature. In addition, no discontinuity (indicating initiation of matrix cracking) in the linear part of the load-deflection curves of these two composites was observed, as is shown in Fig. 1(a-c). However, for SiC/M1a hot-pressed for 0.5 h, a pronounced discontinuity was detected in the load-deflection curve (Fig. 1d), indicating that matrix cracking had occurred. This appeared on all the individual samples tested. A marked change in the crack spacing in this composite was measured, from  $867 \pm 51 \mu\text{m}$  before the test to  $213 \pm 95 \mu\text{m}$  after the test, which was not

shown by the other composites. The average crack spacings in the polished surfaces before the three point bending test, ( $\bar{x}$ ) and in the fractured specimens after the test, ( $\bar{x}$ ) are given in Table III.

## 4. Discussion

### 4.1. Strengths and ultimate failure strains

In comparison with the bulk unreinforced matrix materials, all the composites demonstrated an enhanced flexural strength and a change in their fracture behaviour. The flexural strength values of these materials correlate directly with the quality of the interfacial bond strength. As expected those materials with a weak interface exhibited tough behaviour (Table I). However, there is a discrepancy between the experimental values and the theoretical predictions based on the mixture laws (Table I). The experimental values of strength are lower than the theoretical predictions even assuming that there are no contributions from the matrices. Thus, the reduction in the fibre strength after composite consolidation needs to be considered.

When exposed to high temperatures, carbon fibres oxidize, and hence the composition and structure of

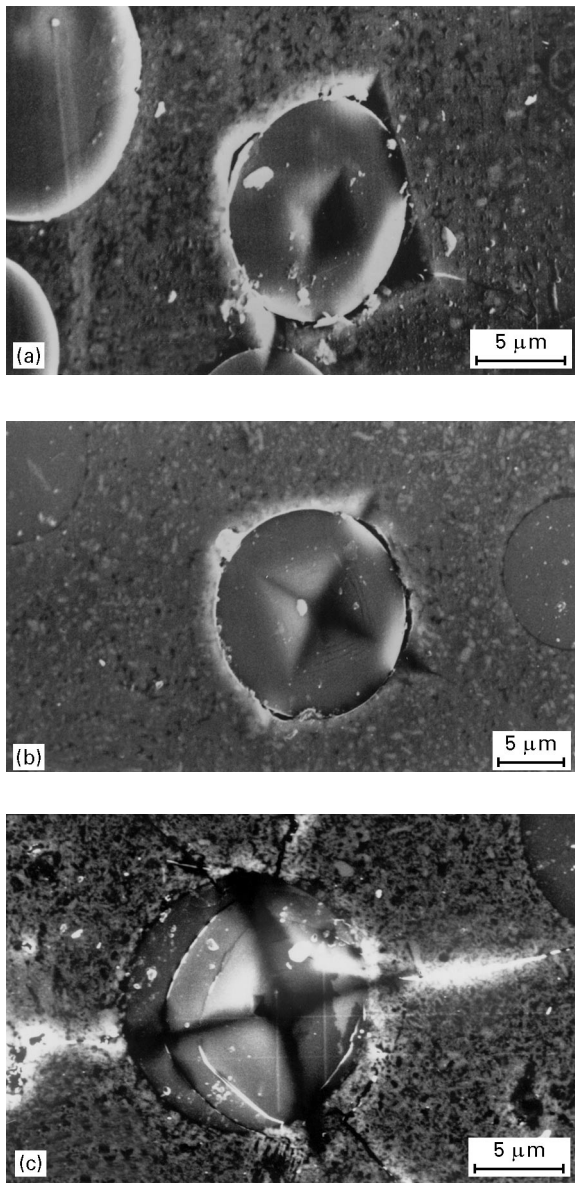
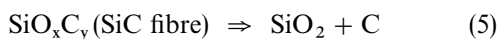


Figure 3 SEM of indentations using the fibre push in test. (a) mullite matrix Nicalon SiC fibre reinforced composite, SiC/M1a hot-pressed at 1550 °C for 0.5 h at 15 MPa, (b) mullite matrix Nicalon-SiC fibre reinforced composite, SiC/M1b hot-pressed at 1550 °C for 1.5 h at 15 MPa and (c) the alumina-silica mixture matrix Nicalon-SiC fibre reinforced composite, SiC/(A + S) hot-pressed at 1400 °C for 0.5 h at 18 MPa.

the SiC fibres changes. Consequently the retained strength of the fibres is affected and also the strength of the composite. Nicalon-SiC fibres generally show reaction even in a reducing atmosphere [15–20] as follows



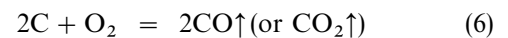
The reduction in strength above 1000 °C is caused by microcrystallization of the fibre structure [15], although the degradation can be alleviated by employing reducing atmospheres [16]. Both a silica phase and the presence of carbon have been detected in the interfacial regions within the SiC/M1a, SiC/M1b and SiC/(A + S) composites using energy dispersive spectroscopy EDS [1, 13], which are believed to be the products of the above reaction. A severe reduction in

TABLE II Interfacial properties of the composites

Matrix starting material	$\alpha$ -alumina/Ludox silica	$\alpha$ -alumina/Ludox silica	$\alpha$ -alumina/Ludox silica
Hot pressing conditions	1550 °C, 15 MPa, 0.5 h	1550 °C, 15 MPa, 1.5 h	1400 °C, 18 MPa, 0.5 h
Fibre/matrix	SiC/M1a	SiC/M1b	SiC/(A + S)
$V_f$ ( $\pm 2\%$ )	60	32	32
$\tau$ (MPa)	$4.4 \pm 2.6$	$29.0 \pm 4.6$	*

\* too strong to be measured

the strength of the SiC composites because of the increase in the processing time at high temperatures was also observed to occur. This can be comprehended by comparing the strength of the SiC/M1a composite hot-pressed for 0.5 h ( $630 \pm 98$  MPa, 40% of the calculated theoretical strength) with that of the SiC/M1b composite hot-pressed for 1.5 h ( $203 \pm 74$  MPa, 22% of the calculated theoretical strength). The results also indicate that the presence of carbon in the interfacial regions does not necessarily lead to a weaker bonded interface, since some composites e.g., SiC/(A + S) had a strongly bonded interface but carbon was still detected in their interfacial regions. This may be because the amount of carbon is reduced owing to a sub-reaction during high temperature processing:



The influence of processing temperature on failure strain may be studied by comparing the carbon fibre/mullite composites, C/M1 and C/M2; the former consolidated at 1550 °C, the latter at 1300 °C. The failure strain of C/M2 ( $0.45 \pm 0.14\%$ ) was higher than that of C/M1 ( $0.37 \pm 0.13\%$ ). The difference may be related to the processing temperatures. Generally the retained strength of carbon fibre can be seriously affected by exposure to high temperatures. However, the strength of C/M2 was significantly lower than that of C/M1 probably because of the lower matrix densification of the former.

#### 4.2. Interfacial strengths, fracture behaviour and fibre pull-out

The presence of a glassy silica phase in the interfacial regions between the SiC fibres and matrices of the SiC/M1a, SiC/M1b and SiC/(A + S) composite was observed by transmission electron microscopy (TEM) and confirmed by EDS in Part I of this work [1]. Silica is expected to form on the fibre surface as a product of the reaction shown in Equation 5. A silica phase is also present in the A + S matrix, resulting in strong interfacial bonding. This is believed to be the main reason that the SiC/(A + S) composite hot-pressed at 1400 °C had strong interfacial bonding, which resulted in the brittle fracture behaviour with no fibre pull-out (Fig. 2e). The strong interface was evidenced when a load was applied to the fibre cross-section. Fracture occurred in the fibre and cracks propagated into the matrix (Fig. 3c).

TABLE III Crack spacings in the composites

Hot-pressing conditions	1550 °C, 15 MPa, 0.5 h	1300 °C, 15 MPa, 40 min	1300 °C, 25 MPa, 1.0 h	1550 °C, 15 MPa, 0.5 h	1550 °C, 15 MPa, 1.5 h	1400 °C, 18 MPa, 0.5 h
Fibre/matrix	C/M1	C/M2	C/(A + S)	SiC/M1a	SiC/M1b	SiC/(A + S)
$V_f$ ( $\pm$ 2%)	52	48	40	60	32	32
$\bar{x}$ ( $\mu$ m)	$106 \pm 19$	$106 \pm 19$	$103 \pm 28$	$867 \pm 51$	–	–
$\bar{x}$ ( $\mu$ m)	No change	No change	No change	$213 \pm 95$	Brittle	Brittle

A weak interface was observed for the SiC fibre/mullite composite, SiC/M1a composite hot-pressed for 0.5 h as shown by the extensive fibre pull-out after the bending test (Fig. 2c) and from the indentation test (Fig. 3a). In this case the interfacial glass layer between the fibres and matrix in the composite may not be sufficiently thick to form a strong bond. The interfacial strength was  $4.4 \pm 2.6$  MPa (Table II). However, a stronger bond between the fibres and matrix in the SiC/M1b hot-pressed for 1.5 h was indicated by the brittle fracture with some short fibre pull-out from the bending test (Fig. 2d) and from the indentation result (Fig. 3b). The interfacial strength was correspondingly higher ( $29.0 \pm 4.6$  MPa). Evidence for a thickened glass interface after longer processing at higher temperatures has been shown elsewhere [20].

The influence of the porosity of the composites on the nature of the interface should also be considered. In the SEM and TEM observations reported in Part I of this work [1], pores and crevices were detected along the interfacial region between the fibres and the mullite matrix within C/M1. These pores may have resulted partly from incomplete densification and partly from the volume change during the mullitization in the matrix. The crevices may have formed as a result of a relative displacement between fibre and matrix on cooling from high temperature because of thermal expansion mismatch. These pores and crevices could contribute to the weak interfacial bonding, resulting in extensive fibre pull-out on failure (Fig. 2a) with a substantial failure strain of  $0.37 \pm 0.13\%$ . However, in the case of the C/(A + S) composite, no such pores or crevices were observed, see Part I [1]. In this case, the magnitude of the fibre pull-out was relatively less (Fig. 2b) with a relatively small failure strain of  $0.16 \pm 0.03\%$ .

#### 4.3. Elastic modulus and its standard deviation

From Table I, the experimental values of the flexural moduli of the carbon fibre/mullite composites, C/M1 and C/M2 are lower than the calculated theoretical values (assuming no matrix contribution) and their standard deviations are high. However, the modulus of the SiC fibre/mullite composite, SiC/M1a hot-pressed at 1550 °C for 0.5 h is relatively high and the standard deviation is low. The density of matrix cracks is considered to be important in this case. The C/M1 and C/M2 composites have a high density of matrix cracks, (see Table II) indicating a small (or zero) con-

tribution from the matrix to the moduli and a high standard deviation in testing. However, SiC/M1a hot pressed for 0.5 h is relatively crack-free (Table II) possibly indicating a significant matrix contribution to the modulus and a low standard deviation in this case.

### 5. Summary and conclusions

Using the three point bending test, the properties of the composites described in Part I of this work were determined. The highest flexural strength of  $737 \pm 128$  MPa, with the highest work of fracture,  $482 \pm 122$  kJ m<sup>-2</sup>, and ultimate failure strain of  $0.37 \pm 0.13\%$  were achieved for the carbon fibre reinforced mullite matrix composite, C/M1, hot pressed at 1550 °C for 0.5 h at 15 MPa. The carbon fibre reinforced composite with a matrix composed of  $\alpha$ -alumina particles dispersed in a silica glass phase, C/(A + S), hot pressed at the lower temperature of 1300 °C had similar  $\sigma$ ,  $E$  and  $W$  values to the C/M1 composite. The SiC fibre mullite matrix composite, SiC/M1a, hot pressed at 1550 °C for 0.5 h also had a high  $\sigma$  value of  $630 \pm 98$  MPa, the  $W$  value of  $293 \pm 111$  kJ m<sup>-2</sup> and an  $\epsilon$  value of  $0.3 \pm 0.1\%$ . However, increasing the hot pressing time to 1.5 h at 1550 °C led to a marked deterioration in the strength value to  $203 \pm 74$  MPa, the  $W$  value to  $43 \pm 25$  kJ m<sup>-2</sup>, and the  $\epsilon$  value to  $0.06 \pm 0.02\%$  and a change from non brittle fracture behaviour to brittle. The carbon fibre mullite matrix prepared from boehmite and Ludox by method B in Part I of this work and hot-pressed at only 1300 °C, to produce a relatively low overall densification (only 84%), still retained a relatively high strength value of  $428 \pm 79$  MPa, a  $W$  value of  $350 \pm 90$  kJ m<sup>-2</sup> and an  $\epsilon$  value of  $0.45 \pm 0.14\%$ .

The fibre reinforcement of the ceramic matrices is highly effective. However, the experimental values of the fracture strength are still lower than the theoretical predictions based on the mixture laws, even assuming that there are no contributions from the matrices. The reduction in strength is attributed to the partial degradation of fibres during composite consolidation at high temperatures during which the carbon fibres may partially oxidize, and thus their composition and structure may change. Consequently the retained strengths of the fibres are affected and also the strengths of the composites.

The interfacial bonding strengths between fibre and matrix in the composites were characterized by the

fibre push in test. Repeated observations showed that the interfacial bonding is weaker in the SiC/M1a composite hot-pressed at 1550 °C for 0.5 h compared with the SiC/M1b composite hot-pressed for 1.5 h. The interfacial strength of the former was  $4.4 \pm 2.6$  MPa as compared to  $29.0 \pm 4.6$  MPa for the latter. However, for the SiC/(A + S) composite, even under a higher load the fibre section was not pushed into the matrix. In addition the fibre fractured and cracks propagated into the matrix, as a result of the strong interfacial bond in this case.

Pores and crevices observed along the interface in the C/M1 composites are believed to have contributed to a reduction in the interfacial strength, leading to extensive fibre pull out during the three point bending test. The amount of silica phase along the interface between the SiC fibre and matrix is thought to be the main factor controlling the interfacial bonding strength in the SiC fibre composites. Substantial silica formation was observed in the interfacial region in the SiC/(A + S) composite hot-pressed at 1400 °C, and a strong bonding was indicated by the indentation results and the fracture surfaces. The thin silica layer at the interface as shown by TEM and EDS for the SiC/M1 composite hot pressed at 1550 °C for 0.5 h, may account for the relatively weak interfacial bonding observed from the indentation results.

The carbon fibre/mullite composites C/M1 and C/M2, that contained high crack densities, showed relatively low elastic moduli and high standard deviations ( $127 \pm 64$  GPa and  $82 \pm 39$  GPa respectively). However, the SiC fibre/mullite composite, SiC/M1a, with a low crack density, showed a relatively high modulus and small standard deviation ( $208 \pm 11$  GPa).

## References

1. WU, F. R. JONES and P. F. JAMES, *J. Mater. Sci.* (in press).
2. R. W. RICE, *Ceram. Engng Sci. Proc.* **7-8** (1985) 589.
3. R. J. KERANS, R. S. HAY and N. J. PAGANO, *Amer. Ceram. Soc. Bull.* **8** (1989) 429.
4. M. D. THOULESS, O. SBAIZERO, L. S. SIGL and A. G. EVANS, *J. Amer. Ceram. Soc.* **72** (1989) 525.
5. L. A. BONNEY and R. F. COOPER, *ibid.* **73** (1990) 2916.
6. E. MOUCHON and PH. COLOMBAN, *Composites* **26** (1995) 175.
7. L. M. SHEPPARD, *Amer. Ceram. Soc. Bull.* **71** (1992) 617.
8. British standard methods of testing, "Method 1005, determination of flexural properties-three point method", BS 2782: Part 10: Method 1005, EN63 (British Standards Institution, 1977).
9. A. KELLY, "Strong solids," 2nd Edn (Clarendon Press, Oxford, 1973).
10. D. J. HANNANT, "Fibre cement and fibre concretes", (Wiley, Chichester, 1978).
11. J. AVESTON, R. A. MERCER and J. M. SILLWOOD, in Proc. Conf. on Composites, Standards, Testings and Design, April 1974, National Physical Laboratory (1974) pp. 93-103.
12. J. WU, M. CHEN, F. R. JONES and P. F. JAMES, *J. Non-Cryst. Solids* **162** (1993) 197.
13. J. WU, PhD thesis, University of Sheffield (1995).
14. D.B. MARSHALL, *J. Amer. Ceram. Soc.* (1984) C-259.
15. G. SIMON and A. R. BUNSELL, *J. Mater. Sci.* **19** (1984) 3658.
16. B. A. BENDER, J. S. WALLACE and D. J. SCHRODT, *ibid.* **26** (1991) 970.
17. T. J. CLARK, R. M. ARONS, J. B. STAMATOFF and J. RABE, *Ceram. Engng Sci. Proc.* **6** (1985) 576.
18. G. ERVIN, Jr., *J. Amer. Ceram. Soc.* **41** (1958) 347.
19. D. W. SHIN, K. H. AUH and K. M. KNOWLES, *J. Ceram. Soc. Jpn* **103** (1995) 311.
20. E. BISCHOFF, M. RÜHLE, O. SBAIZERO and A. G. EVANS, *J. Amer. Ceram. Soc.* **72** (1989) 741.

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