

Continuous fibre reinforced mullite matrix composites by sol–gel processing

Part I *Fabrication and microstructures*

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The fabrication of ceramic composites based on a mullite matrix uniaxially reinforced with either high strength carbon or Nicalon-SiC continuous fibres is described. Sols of high solids contents of up to 40 vol% were prepared using colloidal silica sol with either α -alumina powder or colloidal boehmite as reactants for the preparation of the mullite matrix, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$. Single-stage infiltration was used for the preparation of prepregs for laminating into composites. The composites were consolidated by uniaxially hot-pressing at 1300 or 1550 °C, depending on the matrix precursors and the final phases in the matrix. Highly densified composites with fibre volume fractions of 32–60% were obtained and the microstructures characterised by mercury porosimetry, X-ray diffraction, scanning electron microscopy and transmission electron microscopy.

1. Introduction

Continuous fibre reinforcement has been extensively investigated for the improvement of the strength and toughness of glasses and ceramics [1–7]. Densification of the ceramic matrix and control of the interface between fibre and matrix are vital in the production of composites with useful properties [8, 9]. Mullite combines excellent strength and creep resistance at high temperatures with good chemical and thermal stability and a low thermal expansion coefficient of $4.5\text{--}5.6 \times 10^{-6} \text{ K}^{-1}$ [10]. Therefore, mullite-containing composites have a potential for structural applications at high temperatures [3, 11–13]. However, high processing temperatures in excess of 1700 °C and a lengthy sintering time are required for the preparation of monolithic mullite by the traditional sintering route using micron-scale starting materials [10]. A lower mullitization temperature of 980 °C can be achieved by molecular-scale mixing of alkoxide precursors. However, the resultant sols have low solid concentrations [14, 15]. In addition, although a sol produced from alkoxide precursors can be used for the fabrication of a continuous fibre reinforced composite [8, 16], multiple-impregnation is required. The solids content of sols can be enhanced by addition of fillers by mixing different size powers in one dispersion [17]. To achieve either monolithic mullite or mullite matrix composites with a high density at the lowest possible processing temperatures, the development of a novel technique is needed, which has the advantages of the sol–gel route but none of the disadvantages. In previous work [18, 19] it was shown that a sol containing 40 vol% solids, of the mullite composition, $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, could be prepared using α -alumina

powder and colloidal silica sol. This sol was transformed into a gel from which a mullite monolith was obtained by pressureless sintering at 1600 °C for 2 h. A density of $3.10 \times 10^3 \text{ kg m}^{-3}$ for the bulk sample was obtained, which corresponds to 98% of the theoretical density of $3.17 \times 10^3 \text{ kg m}^{-3}$ for mullite. The microstructures of the bulk materials have been characterized and the densification mechanism analysed [20]. It was demonstrated that a unidirectional carbon fibre reinforced crystalline mullite matrix composite could be fabricated by single-stage infiltration with this sol and final consolidation by hot-pressing [19].

In this paper, the fabrication of composites using this sol–gel technique is described, in which either high strength carbon or Nicalon silicon carbide fibres were employed as reinforcements. The matrices were either mullite or an alumina-silica stoichiometric mixture. Another novel technique is also reported for the preparation of bulk mullite specimens and mullite matrix fibre reinforced composites using colloidal boehmite instead of α -alumina powder as an alumina source via lower mullitisation temperature of 1300 °C. The microstructures of the composites were characterized by X-ray diffraction (XRD), scanning electron microscopy (SEM) and transmission electron microscopy (TEM). The fibre–matrix interfaces of the composites were examined by TEM. The flexural strengths and fracture behaviour of the composites are discussed in detail in part 2 of the present paper.

2. Experimental procedure

Prepregs of the unidirectional fibre reinforced ceramic matrix composite samples were prepared by the

single-stage infiltration of the fibre tows with a colloidal sol corresponding to an oxide composition of $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$. Two methods, A and B, were employed for the preparation of the sols: method A, which is described elsewhere [19], used Ludox colloidal aqueous silica sol (Dupont AS40, 40 wt % of silica particles of 5–50 nm in diameter) and α -alumina powder (Alcoa A1000 SG, with an average size of 0.5 μm) as starting materials. Method B employed a colloidal boehmite (GC Powder, 20 nm in size) instead of α -alumina.

Briefly, in method A, the as-received α -alumina powder was dispersed in the Ludox colloidal silica sol to produce a homogenous sol. The viscosity of the sol was sufficiently low for the efficient impregnation of the fibre tow. The high solids concentration (alumina-silica mixture) of 40 vol % (68 wt %) enabled single-stage infiltration to be employed for the formation of a dense matrix.

In method B, also described in [21], the Ludox silica sol was diluted with distilled water. The pH was then adjusted to between 2–3 by adding dilute HCl and maintained at this value while boehmite powder was dispersed in the sol with the help of a magnetic stirrer. A highly stable sol, corresponding to an oxide composition of $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$, could be prepared in this way. Gelation of the sol was allowed to occur by heating at 90 °C. After calcination at 500 °C for 2 h, the porous gel was crushed and ground into a powder with a particle size of 38 μm using a mortar and pestle. The powder was redispersed in distilled water by means of ball-milling to produce sols with controlled solids concentrations ranging from 30–50 vol %. Ball-milling was carried out for 24 h in a sealed plastic chamber containing half the volume of ZrO_2 balls of 9 mm in diameter. The sols prepared in this way could be converted to gels on drying in an oven at 90 °C. Monolithic mullite bulk samples with 98% of the theoretical density could be obtained by pressureless-sintering of the gels at 1300 °C for 2 h. The sol from the redispersion, with a 40 vol % solids concentration, was used for the fabrication of fibre reinforced composites by single-stage infiltration.

Grafil LXA unsized high strength continuous carbon fibre tow has a diameter of 8 μm , strength of

4 GPa, an elastic modulus of 224 GPa, an ultimate strain of 1.78% and an average density of $1.81 \times 10^3 \text{ kg m}^{-3}$. The Nicalon-207 silicon carbide is a polyvinyl alcohol sized continuous fibre yarn which has a strength of 2.6 GPa, an elastic modulus of 189 GPa, an ultimate strain of 1.37%, an average density of $2.55 \times 10^3 \text{ kg m}^{-3}$ and a diameter of 14 μm . Nicalon-207 SiC polyvinyl alcohol sized fibre yarn was de-sized by passing through hot water at 90 °C, and then stored in vacuum ready for the preparation of the composites.

Unidirectional fibre reinforced composite prepreps were fabricated by infiltrating the continuous fibres using the sols without additional binder. The fibre tow was impregnated in a single-stage by passing it through the sols and wound onto a hexagonal drum to form a preimpregnated sheet prepreg of 10 mm in thickness. The prepreg was dried in air at room temperature for 48 h and in an oven at 90 °C for a further 48 h. By careful control of the solids concentration, composites with a fibre volume fraction in the range of 30–60% could be prepared. The sol prepared by method A was used for the fabrication of the carbon fibre and Nicalon-SiC fibre composite prepreps and the sol prepared by method B was only used with carbon fibres.

The dried composite prepreg was cut into circular discs of 40 or 50 mm in diameter to fit a graphite die. In the graphite die assembly, the composite stack was sealed by two thick layers of graphite powder to ensure that a reducing atmosphere of CO existed during hot-pressing. The uniaxial hot-pressing conditions depended on starting materials and final product (see Table I). In Table I A + S refers to the matrices consisting of α -alumina particles dispersed in silica glass, M1 and M2 refer to crystalline mullite matrices produced by methods A and B respectively. The composite samples prepared by method A were consolidated by hot-pressing at 1300 °C, for 1 h at a 25 MPa pressure or at 1400 °C, for 0.5 h at 18 MPa for the production of the alumina-silica 'mixture' matrix (A + S) composites reinforced by carbon fibre and silicon carbide fibre respectively, and at 1550 °C, for 0.5 or 1.5 h at 15 MPa for the formation of the mullite matrix (M1) composites reinforced by carbon fibre or

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

Matrix starting material	α -alumina/ Ludox silica	α -alumina/ Ludox silica	α -alumina/ Ludox silica	α -alumina/ Ludox silica	α -alumina/ Ludox silica	Boehmite/ Ludox silica
Hot-pressing conditions	1300 °C 1 h 25 MPa	1400 °C 0.5 h 18 MPa	1550 °C 0.5 h 15 MPa	1550 °C 0.5 h 15 MPa	1550 °C 1.5 h 15 MPa	1300 °C 40 min 15 MPa
Fibre/matrix	C/(A + S)	SiC/(A + S)	C/M1	SiC/M1a	SiC/M1b	C/M2
V_f ($\pm 2\%$)	40	32	52	60	32	48
ρ_c/ρ_{0c} 100%	98	98	95	98	99	84

(A + S): a matrix composed of α -alumina particles dispersed in a densified silica host with $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ composition; M1: a mullite matrix produced from α -alumina powder and Ludox colloidal silica sol; M2: a mullite matrix produced from boehmite gel powder and Ludox colloidal silica sol; \pm : standard deviation; ρ_c : measured density of composite; ρ_{0c} : calculated theoretical density (see text); V_f : volume fraction of fibres in composite.

silicon carbide fibre. Composites prepared by method B were consolidated by hot-pressing at 1300 °C, for 40 min at 15 MPa for the preparation of the mullite matrix (M2) composite reinforced by carbon fibre. All hot-pressed composite samples were allowed to cool naturally to room temperature overnight within the furnace.

The densities of the composite samples ρ_c were determined by mercury porosimetry using a Micromeritics Pore Size 9320 with a pressure range of ~ 174 MPa. The volume fraction of fibres in the composites and the crack spacing in the polished surface of the composites were determined by computer-controlled image analysis using an AMS Optomax V. The volume fractions of carbon fibres in the composites were confirmed from the weight loss after firing at 1000 °C for 3 h in air. After this treatment, the remaining oxide matrix was white in appearance on the outside surface and on the inside after fracture, indicating that no carbon remained. The maximum theoretical density of each composite (assuming no porosity), ρ_{0c} was calculated using the following equation:

$$\rho_{0c} = V_m \rho_m + V_f \rho_f \quad (1)$$

where ρ_m is the theoretical density of the matrix; ρ_f is the theoretical density of the fibre; V_f is the measured volume fraction of fibre in the composite; and V_m is the measured volume fraction of matrix in the composite. The phases present in the matrix of the composites were identified by X-ray diffraction using a Philips 1710 X-ray Diffractometer with a scanning speed of 2° (2θ) min^{-1} . Scanning electron microscopy (SEM) was performed on a Jeol JSM6400. Transmission electron microscopy (TEM) and Energy dispersive spectroscopy (EDS) were performed on a Philips EM400T. Thin sections for TEM were prepared by ion beam milling a specimen which had been mechanically dimpled to less than 30 μm in thickness using fine grinding paste.

The thermal expansion coefficients of the matrix materials were measured using a dilatometer with fused silica as a reference [21]. A bulk sample of the $3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$ "mixture", i.e., α -alumina particles dispersed in a silica glass, with a density of $3.23 \times 10^3 \text{ kg m}^{-3}$, was prepared by method A and fired at 1300 °C for 2 h, and the mullite bulk sample with a density of $3.10 \times 10^3 \text{ kg m}^{-3}$ was also prepared by method A and fired at 1600 °C for 2 h. The specimens were cut from the sintered bulk samples, and then, ground and polished to a diameter of 10 mm and length of 30 mm. The thermal expansion coefficients quoted are average values in the temperature range 25–700 °C.

3. Results and discussion

3.1. Microstructures of composites

The microstructures of the C/(A + S) composite in which the matrix consisted of α -alumina particles imbedded in a densified silica glass were described in [20]. Fig. 1 shows the XRD results for two of the hot-pressed composites, pattern (a) corresponds to the carbon fibre composite, C/M1 prepared by method A,

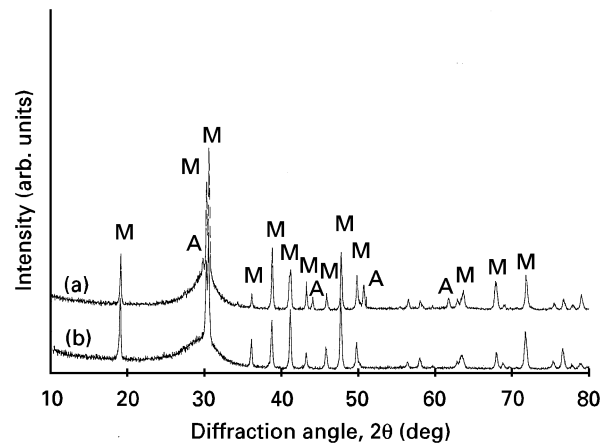


Figure 1 XRD patterns of mullite matrix carbon fibre reinforced composites; (a) C/M1 hot-pressed at 1550 °C for 0.5 h at 15 MPa using α -alumina powder and colloidal silica as matrix starting materials and (b) C/M2 hot-pressed at 1300 °C for 40 min at 15 MPa using boehmite gel powder and colloidal silica as matrix starting materials.

hot-pressed at 1550 °C, for 0.5 h at 15 MPa and pattern (b) to the composite, C/M2 prepared by method B, hot-pressed at 1300 °C for 40 min at 15 MPa. The two composite matrices are predominantly crystalline mullite, although in pattern (a), a trace of unreacted alumina is evident. For the SiC/M1a composite processed by method A, hot pressed under the same conditions as the C/M1 composite, XRD showed a similar result to pattern (a).

Fig. 2a shows a polished cross-section of the C/M1 composite, perpendicular to the fibre direction. The round black dots are the carbon fibre cross-sections, uniformly distributed in the mullite matrix (grey colour). There are parallel areas in which less fibres are observed, which originated from the processing method and indicated the boundaries between fibre tows. Fig. 2b shows a polished section through the C/M1 composite approximately parallel to the fibre direction. A dense mullite matrix with a uniform distribution of carbon fibres ($V_f = 0.52$) was revealed. No large pores were observed, which was consistent with the measured density of 95% of the calculated theoretical density of $2.46 \times 10^3 \text{ kg m}^{-3}$ for the composite. Mechanical polishing resulted in some degree of damage or "pluck-out" at the fibre–matrix interface (white areas indicated by arrows) suggesting that little or no chemical bonding had occurred. At high magnification (Fig. 2c), some small pores ($\sim 3 \mu\text{m}$) or crevices were observed along the interface between the matrix and the fibres. Some regularly spaced microcracks were observed in the matrix, which are perpendicular to the fibre direction in both Fig. 2 (b and c), which are believed to be induced by thermal mismatch between the fibres and mullite matrix. Thermally induced cracks are produced by stresses resulting from a mismatch in the expansion coefficients of the carbon fibres and the mullite matrix on cooling from the high processing temperature to room temperature. The measured thermal expansion coefficient is $4.83 \times 10^{-6} \text{ K}^{-1}$ for the bulk samples of mullite and that of the carbon fibres is about zero [22]. It is

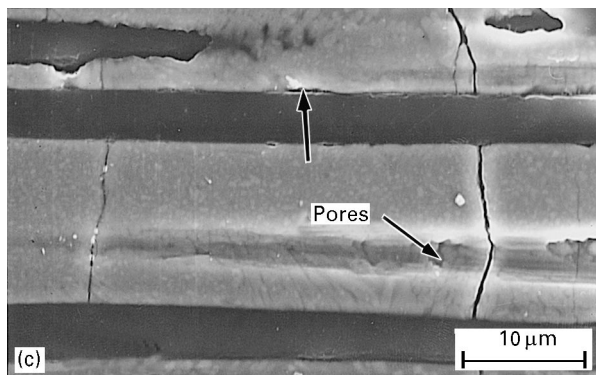
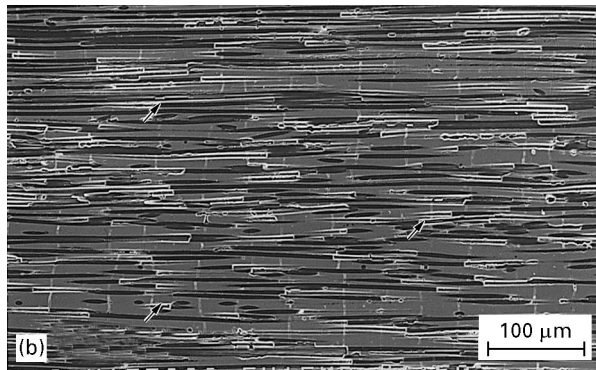
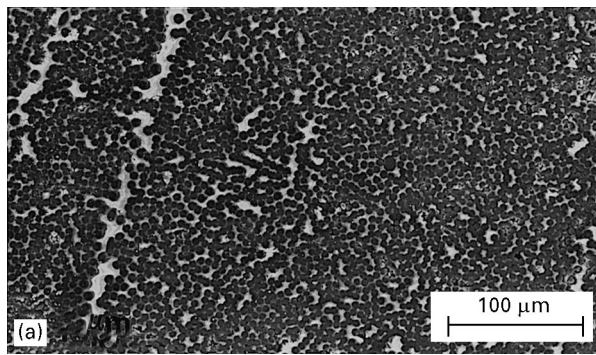


Figure 2 SEM of polished sections through the C/M1 composite hot-pressed at 1550 °C for 0.5 h at 15 MPa using α -alumina powder and colloidal silica as matrix starting materials; (a) cross-section perpendicular to the fibre direction; (b) section parallel to the fibre direction, arrows pointing to mechanical polishing damage (fibre plucked from the matrix); (c) high magnification of Fig. 2b.

estimated that a tensile stress of the order of ~ 700 MPa would be induced in the mullite matrix upon cooling to room temperature from 1550 °C [21]. The magnitude of this stress is significantly higher than the flexural strength of mullite, which has been found to be approximately 150 MPa [21] and would lead to multiple cracking in the matrix. The thermally induced cracks and the interfacial pores are believed to be the main contributors to the 5% porosity within the C/M1 composite.

Fig. 3a shows a polished cross-section through the C/M2 composite, perpendicular to the fibre direction. The black dots are the carbon fibre cross-sections distributed in the mullite matrix (white). Pores in the matrix are more frequently observed than in C/M1, which is in accord with the higher measured porosity (16%) for C/M2. In some cases pores are joined up by

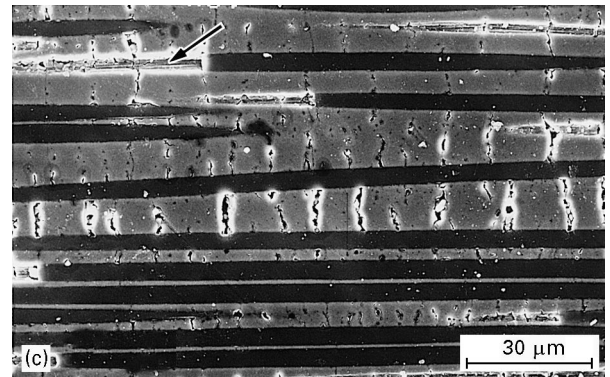
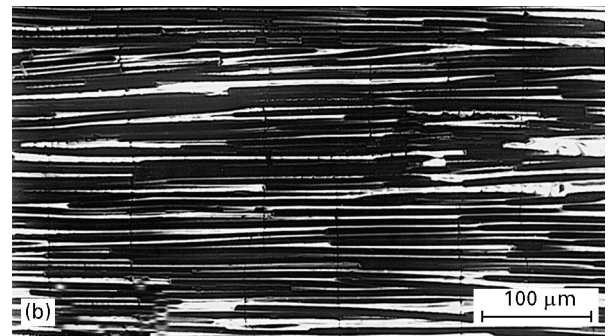
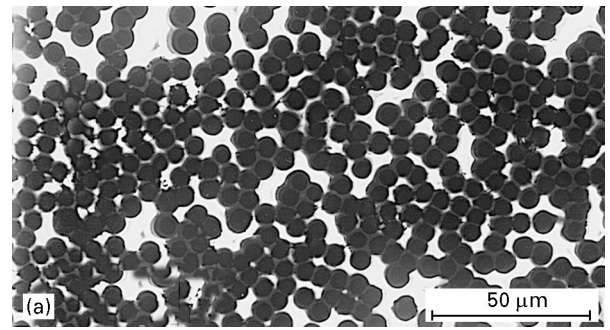


Figure 3 SEM of polished sections through the C/M2 composite hot-pressed at 1300 °C for 40 min at 15 MPa, using boehmite and colloidal silica as matrix precursors, (a) cross-section perpendicular to the fibre direction; (b) parallel to the fibre direction (at lower magnification); (c) parallel to the fibre direction, arrow pointing to mechanical polishing damage (fibre plucked from the matrix).

cracks perpendicular to the fibres. The origin of these pores is not understood but they probably arise from the incomplete densification of C/M2 during hot pressing. Parallel cracks running through the samples are clearly observed at lower magnification (Fig. 3b) and again probably result from the thermal mismatch stresses as in the C/M1 composite. In Fig. 3c some fibre pluck out during polishing was observed (indicated by arrow) again suggesting a lack of chemical bonding at the fibre/matrix interface.

The thermal expansion coefficient for the SiC fibres at $4.0\text{--}5.0 \times 10^{-6} \text{ K}^{-1}$ [22] is similar in magnitude to that of mullite. Thus, the potential for thermal mismatch cracking in the SiC/mullite composites is much lower as suggested by the observation of few cracks in a random polished section through the SiC/M1a composite (Fig. 4a). A polished section perpendicular to the fibre direction (Fig. 4b) shows that some fibres were in contact.

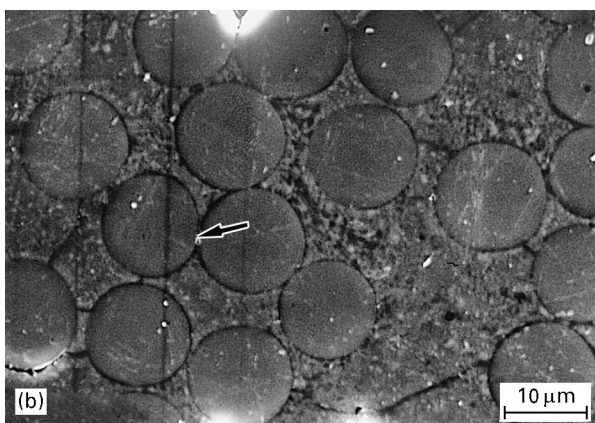


Figure 4 SEM of polished sections through the SiC/M1a composite hot-pressed at 1550 °C for 0.5 h at 15 MPa, using α -alumina powder and colloidal silica as matrix starting materials; (a) random section, arrow pointing to mechanical polishing damage (fibre plucked from the matrix); (b) cross-section perpendicular to the fibre direction, arrow pointing to fibres in contact.

Optical microscopy of the SiC/(A + S) composite ($V_f = 0.32$) revealed a complex distribution of cracks in the matrix with spacing of $\cong 4.0$ mm between the larger cracks and additional fine microcracks (not shown). In addition, some cracks had propagated through the fibres (Fig. 5). There was no evidence for fibre removal during polishing, indicating a strong interfacial bond between the fibre and matrix. The thermal expansion coefficient of the bulk alumina-

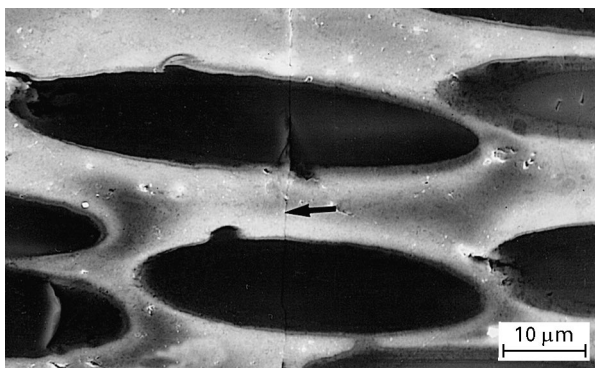


Figure 5 SEM of random polished section through the SiC/(A + S) composite hot-pressed at 1400 °C for 0.5 h at 18 MPa, using α -alumina powder and colloidal silica as matrix starting materials, arrow pointing to a crack through fibres and matrix.

silica (A + S) matrix on its own was measured as $4.0 \times 10^{-6} \text{ K}^{-1}$, which is also similar in magnitude to that of the SiC fibre. However, the A + S matrix is microstructurally non uniform on a sub-micron scale (0.5 μm alumina particles dispersed in silica glass). Locally induced thermal mismatch stresses could occur between the silica ($\alpha \sim 0.5 \times 10^{-6} \text{ K}^{-1}$) and the SiC fibres leading to tensile stresses in the fibres after processing. This might explain the origin of the cracks in the composite. The strong interfacial bond would facilitate the propagation of cracks across the fibres/matrix interface. However this needs to be confirmed.

To confirm that the cracks in the carbon fibre/mullite matrix composites were caused by thermal expansion mismatch induced stresses, a carbon fibre prepreg prepared by method A was heated at 900 °C for 3 h under oxidizing conditions to completely remove the fibres. It was then heated at 1600 °C for 2 h, and finally allowed to cool naturally to room temperature within the furnace. The polished surface of this sample was examined by optical microscopy and no parallel multiple matrix cracks were observed. This indicates that the cracks in the composites were formed by residual stresses arising from constrained contraction on cooling and not as a result of sintering. From previous work, it was also found that cracks induced by sintering in a composite matrix do not generally have a preferred orientation throughout the matrix [17, 21].

3.2. Fibre/matrix interfaces in the composites

The interfaces between the fibre and matrix in the composites were examined by TEM as shown in Figs 6 (a and b) and 7(a-c). A sharply defined interface was observed between the carbon fibre and the mullite matrix in the C/M1 composite (Fig. 6a). Some crevices appear to occur along the interface which would further weaken the interfacial bonding. The crevices are believed to be the consequence of the thermal expansion mismatch between fibre and matrix during cooling. No obvious chemical interaction between fibre and matrix was evident. Some mullite grains near the fibre surface were observed to have grown parallel to the fibre direction. Dislocations were also observed in the mullite grains or along grain boundaries (indicated by arrows). TEM also revealed little sign of interaction between fibre and matrix in the C/(A + S) composite (Fig. 6b), in which the matrix was composed of dispersed α -alumina particles in a densified silica glass host, but no crevices or pores were observed along the interface.

Evidence for a thin continuous interfacial layer between the SiC fibres and the mullite matrix in the SiC/M1a composite was revealed by TEM (Fig. 7a). EDS analysis (Fig. 7b) indicated the layer mainly consisted of silica and carbon (amorphous or crystallized). It is possible that the SiC fibre degraded to form a silica layer at high temperatures. This will be discussed further in our next publication on this topic. In the SiC/(A + S) composite in which the matrix was

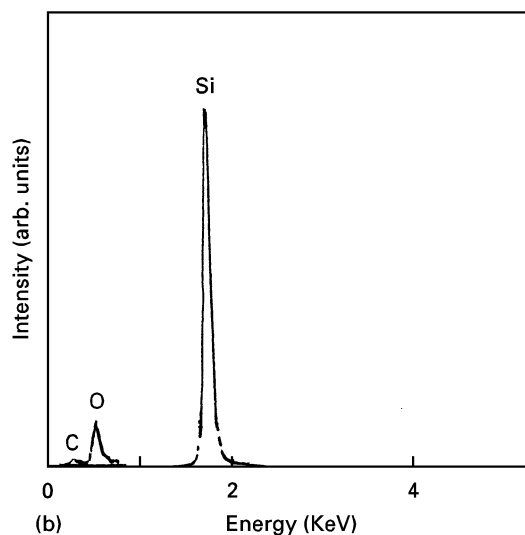
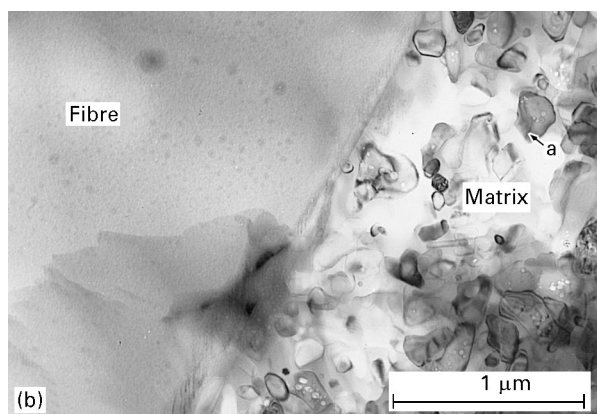
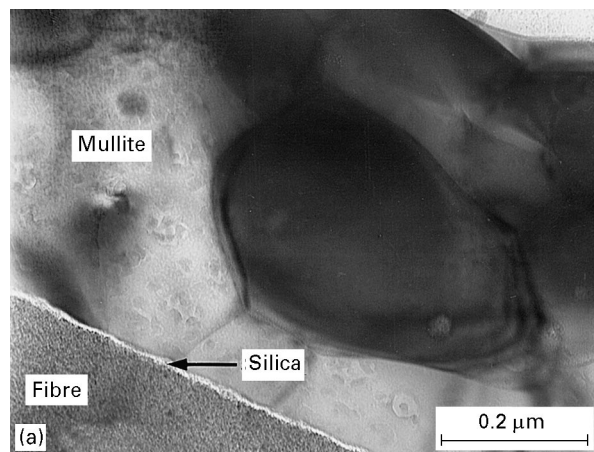
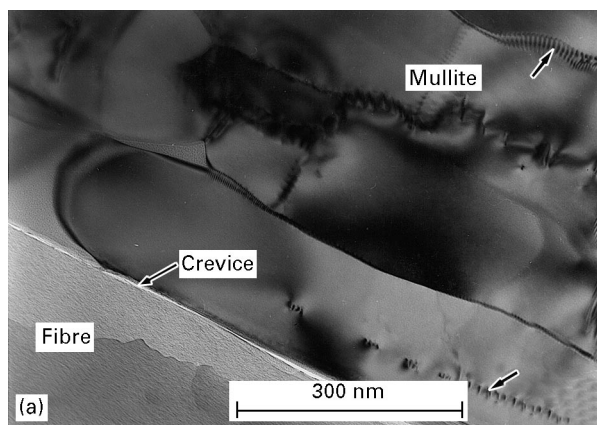


Figure 6 TEM of the interfaces between fibre and matrix for carbon fibre composites; (a) the mullite matrix carbon fibre reinforced composite, C/M1 hot-pressed at 1550 °C for 0.5 h at 15 MPa, arrows pointing to dislocations; and (b) the alumina-silica mixture matrix carbon fibre reinforced composite, C/(A + S) hot-pressed at 1300 °C for 1 h at 25 MPa, **a** denotes an α -alumina particle.

composed of dispersed α -alumina particles in a densified silica glass phase the interface between fibres and matrix appeared less clearly defined (Fig. 7c). EDS analysis indicated that a silica phase was present in the interfacial region [21]. This facilitated relatively strong bond formation between fibre and matrix, confirming the earlier observations.

3.3. Densification and mullitization of composite matrices

Highly densified composites were produced using method A, including C/(A + S), C/M1, and SiC/M1a which have densities ranging from 95–98% of the theoretical density (Table I). In method A glass viscous sintering is the main matrix densification mechanism as discussed previously [20]. Production of the mullite matrix composites can be divided into two steps during hot-pressing. The first step is the densification of the matrix of silica glass containing dispersed α -alumina particles at temperatures from 1200–1400 °C. In previous work TEM showed that the porous glassy host containing α -alumina particles could be fully densified at 1300 °C by viscous sintering even without applying pressure [20]. The next step was mullite formation in the densified matrix at temperatures above 1400 °C by the reaction of α -alumina

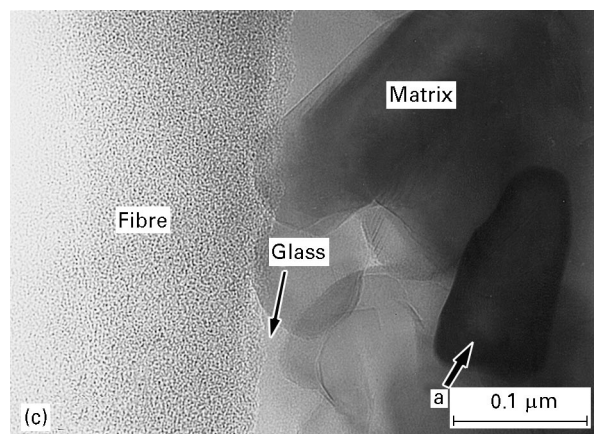


Figure 7 TEM of the interfaces between fibre and matrix for silicon carbide fibre composites; (a) the mullite matrix Nicalon-SiC fibre reinforced composite, SiC/M1a hot-pressed at 1550 °C for 0.5 h at 15 MPa; (b) EDS (intensity as function of energy) of the interfacial layer between fibre and matrix in Fig. 7a; (c) the alumina-silica mixture matrix Nicalon-SiC fibre reinforced composites, SiC/(A + S) hot-pressed at 1400 °C for 0.5 h at 18 MPa, arrow pointing to an α -alumina particle.

particles with the silica matrix. Substantial mullite was formed at temperatures above 1550 °C. The hot-pressing facilitated densification and consolidation during mullitization.

A density, 98% of the calculated theoretical, $2.62 \times 10^3 \text{ kg m}^{-3}$, was achieved for the carbon fibre composite hot-pressed at 1300°C with the (A + S) matrix and a density, 95% of the calculated theoretical of $2.46 \times 10^3 \text{ kg m}^{-3}$ for the carbon fibre mullite matrix composite at 1550°C (Table I). The glass viscous sintering mechanism was fully realized at 1300°C and a densified composite with an alumina-silica matrix was obtained with the aid of external pressure (held for 1 h under 25 MPa). For the carbon and silicon carbide fibre reinforced composites hot-pressed at 1550°C , from XRD and TEM results, substantial mullitization (approximately 90% or more by volume) could be achieved in the matrix.

Complete mullitization was not achieved for hot-pressing at 1550°C because of the slowness of the interdiffusion processes [23]. To study the mullitization process, a further experiment was performed. Two bulk gel samples, prepared by method A, were fired at $20^\circ\text{C min}^{-1}$ to 1600°C , one with no holding time and the other with a holding time of 2 h. XRD showed that the percentage of mullite formed was similar in both cases (approximately 95 vol %). This indicates that the mullitization is initially rapid at temperatures around 1600°C , but as the mullite grains form and grow, the process is slowed down, requiring a longer sintering time or higher temperature for completion. Full mullitization was achieved by firing the bulk sample prepared by method A at 1700°C for 3 h. In this case no unreacted glass phase or alumina were evident, as shown by SEM of a polished surface of the sample which was thermally etched at 1500°C for 2.5 h after sintering (Fig. 8).

The lower mullitization temperature at 1300°C for the mullite matrix composite prepared by method B (Fig. 1b) resulted from the finer starting material, colloidal boehmite, used as the source of alumina. Because of this lower processing temperature, the carbon fibres in the composite should be less damaged by oxidation, and thus should maintain their strength better than that of the fibres in the composites hot-pressed at higher temperatures. Consequently, the toughening effect of the fibres should be enhanced for the lower processing temperature of 1300°C . However, substantial porosity (16%) was found in the matrix, indicating that the processing needs further improvement.

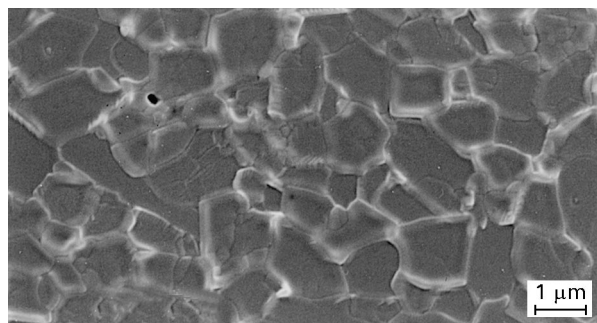


Figure 8 SEM of a polished surface of sample prepared by method A, fired at 1700°C for 3 h and then thermally etched at 1500°C for 2.5 h.

4. Conclusions

Carbon fibre and SiC fibre continuous reinforced mullite matrix composites were successfully fabricated by sol-gel processing, using α -alumina powder and colloidal silica sol as starting materials by single stage infiltration followed by hot-pressing at 1550°C . Highly densified carbon fibre mullite matrix composites (95% of the calculated theoretical density) and silicon carbide fibre mullite matrix composites (98% of the calculated theoretical density) were obtained. Highly densified composites with unmullitized matrices consisting of α -alumina particles in silica glass were also prepared.

Colloidal boehmite was also used as an alumina source to prepare a sol with a high solids content for the fabrication of continuous fibre reinforced mullite matrix composites. This novel processing method for composites resulted in a lower mullitization temperature of 1300°C . The solids content of the sols was up to 40 vol % and the viscosity was sufficiently low for the fabrication of composites using single stage infiltration. However, the composite had a density of only 84% of the theoretical value so that the processing needs further improvement.

The microstructures of the composites were examined by SEM and TEM. Parallel cracks were observed in the carbon fibre reinforced composites, resulting from thermal expansion mismatch. For the C/M1 composite a sharply defined interface with little sign of interaction was observed between carbon fibres and mullite matrix, and some crevices were also observed along the interface. A similar interface was also observed for the C/(A + S) composite but no crevices were observed. A highly densified and nearly crack-free mullite matrix was observed in the SiC/mullite composite, SiC/M1a. For this composite a thin continuous interfacial silica layer was also observed by TEM and EDS, which may result from reaction at the SiC fibre surface at high temperatures. Elongation of the mullite grains near the fibre surface was observed in both of the C/M1 and SiC/M1a composites. A relatively strong interfacial bond was evident in the SiC/(A + S) composite and a complex distribution of cracks was observed. The interface for this composite was less clearly defined and a significant amount of silica phase occurred in the interfacial region. The silica phase could originate from reaction at the SiC fibre surface and from the unmullitized A + S matrix in this case. The differences in the interfaces are attributed to the different starting materials used and different consolidation conditions.

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