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High-strength silicon carbide fibre-reinforced glass-matrix composites

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Silicon carbide fibre-reinforced glass-matrix composites have been fabricated and tested. Two fibre forms, a 140 μm diameter monofilament and a 10 μm diameter filamentary yarn, were incorporated into a matrix of borosilicate glass. The hot-pressing fabrication procedure resulted in fully dense unidirectionally reinforced specimens with excellent flexural strength and fracture toughness over the temperature range 22 to 700° C. In addition, composite thermal expansion was found to be nearly independent of fibre orientation indicating that multiaxially reinforced composites should be readily fabricable without the occurrence of extensive cracking.

1. Introduction

The use of fibre-reinforced resin and metal-matrix composites for structural applications has, during the past 10 to 15 years, increased to the point where many of these systems, such as boron and graphite-reinforced epoxy and boron-reinforced aluminium, are now incorporated in commercially available products that range from advanced jet aircraft to sporting goods. The use of these composite materials has, in almost all cases, been restricted to applications that experience a maximum use temperature of no more than 300° C. During the past few years, however, the interest in composite materials that could extend this temperature capability has been stimulated by the results of several research programmes dealing with glass-matrix composites reinforced with either graphite [1–4] or alumina fibres [5]. In all of these programmes it was found that the use of fibres exhibiting high strength and stiffness was successful in reinforcing lower modulus glass matrices. The graphite fibre-reinforced glass system demonstrated exceptionally high levels of strength, fatigue resistance and fracture toughness while also being susceptible to fibre oxidation during elevated temperature exposure. In contrast, the alumina fibre-reinforced silica-matrix composite [5] was unaffected by exposure to temperatures above 1000° C in air; however, the overall levels of strength and toughness attained were far less than those of the graphite-reinforced glass system. The silicon carbide fibre-reinforced glass-matrix composites to be described here provide a unique combination of both high levels of mechanical performance along with excellent oxidation resistance.

There have recently become available on the commercial market two new types of high-temperature fibres based on silicon carbide. A SiC monofilament of 140 μm diameter, that is fabricated by chemical vapour deposition onto a carbon filament core, is now being produced in pilot plant quantities by the AVCO Systems Division, Lowell, Mass. This fibre exhibits an average tensile strength of up to 3450 MPa, has a temperature capability of over 1300° C, and is stable in oxidizing environments. It has a density of 3.2 g cm^{-3} and an elastic modulus of 415 GPa.

The second type of SiC fibre, recently synthesized by Yajima *et al.* [6] in Japan, consists of continuous length SiC yarn that is produced from an organo-metallic polymer. The tows of yarn contain ~ 2000 fibres/tow with average fibre diameters of 10 μm. The SiC fibre is highly flexible with an extremely smooth surface. The average strength of this fibre, as measured by the authors, is 2060 MPa with the fracture surfaces of the highest strength fibres indicating a dependence on surface flaws. Tensile strengths of up to 3450 MPa have been reported for this fibre by the

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manufacturer with a use temperature of up to 1300° C. With its physical similarity to graphite yarn, this fibre can be incorporated into glass matrices by using the processes already developed for graphite–glass composites. The SiC yarn density is approximately 2.7 g cm^{-3} and the elastic modulus is 221 GPa.

During the present study, it has been found that by incorporating these SiC fibres into a borosilicate glass matrix, a high strength, low density, high modulus composite material with considerable fracture toughness that can retain its properties to temperatures over 600° C, can be obtained.

2. Composite fabrication

The steps in the fabrication of a SiC monofilament-reinforced glass composite were as follows. The SiC fibre was wound with the desired spacing on a drum, bonded together at periodic intervals with polystyrene, cut into individual tapes and then stacked up in the hot-pressing die to form the composite by alternating an SiC tape with glass powder. The amount of powder was varied to make two types of composite samples; one with 65 vol% SiC and one with 35 vol% SiC fibre. The composite lay-up was then hot-pressed for 20 min at a maximum temperature of 1150° C and pressure of 6.9 MPa in an argon atmosphere. It was found that this hot-pressing schedule resulted in complete densification of the 7740* glass with very little bubble or void formation. A cross-section of a typical 65 vol% SiC composite is shown in Fig. 1.

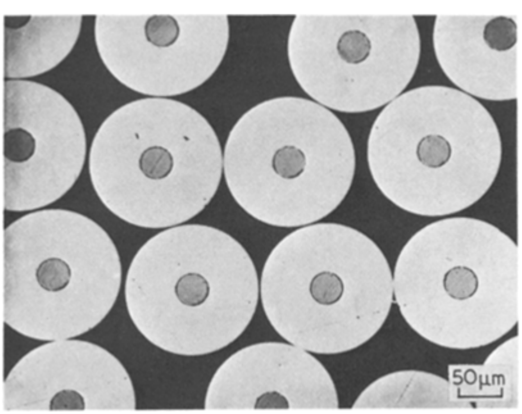

Figure 1 Cross-section of 65 vol % SiC monofilament-reinforced 7740 borosilicate glass composite.

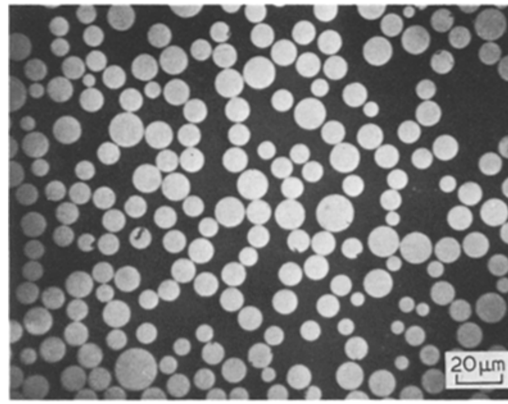

Figure 2 Cross-section of 40 vol% SiC yarn-reinforced 7740 borosilicate glass composite.

The SiC yarn-reinforced 7740 glass specimens were fabricated using the identical procedure developed for the fabrication of graphite fibre-reinforced glass-matrix composites [3, 4]. The SiC yarn was passed through a slurry of glass powder and isopropyl alcohol, dried, and then cut into tapes of the appropriate length to fit the hot-pressing die. Sufficient tape was then stacked in the die to obtain the desired thickness composite and hot-pressed in vacuum for 1 h at 1200° C, 14 MPa. This hot-pressing schedule was found to result in void-free composites with complete densification of the glass matrix. A cross-section of a typical 40 vol% SiC composite is shown in Fig. 2.

3. Composite properties

3.1. Flexural strength

Three-point bend tests of both 35 vol% and 65 vol% monofilament-reinforced SiC-7740 composites were performed at temperatures from 22 to 700° C in air. The maximum flexural tensile stress was calculated at fracture using simple elastic beam theory and the resultant data are presented in Fig. 3. The 65 vol% SiC/7740 composites exhibited an average 22° C strength of 830 MPa which increased to 930 MPa at 350° C and then dramatically increased to 1240 MPa at 600° C. The reason for the increase in strength with increasing temperature is that the viscosity of the glass matrix is decreasing. At 700° C the effective flexural strength of the composite drops off due to the fact that the matrix is now deforming completely plastically with the result

*7740 is the Corning Glass Works designation for a borosilicate glass.

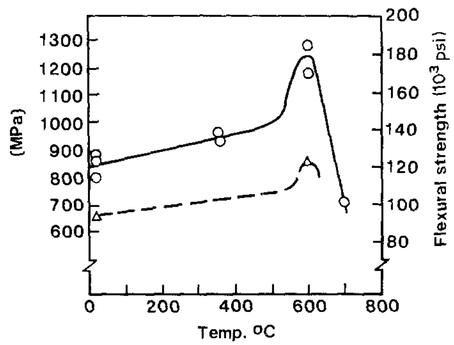

Figure 3 Flexural strength, determined by three-point bend in air, as a function of test temperature for SiC monofilament-reinforced 7740 borosilicate glass. ○, 65 vol% fibre; △, 35 vol% fibre.

that the composite does not fracture, but instead is bent into a "U" shape. Because of this large deformation the value of strength calculated is inaccurate; however, it does express the major change in load-bearing capacity of the composite. Composite samples with 35 vol% SiC fibres act in an identical manner except fracture occurs at lower stress levels.

The data obtained by the three-point bend testing of SiC yarn-reinforced 7740 composites are presented in Fig. 4. In a fashion similar to that of SiC monofilament/7740 composites, the flexural strength increased with increasing temperature due to the increased strain capacity of the glass matrix. The values increased from an average of 290 MPa at 22° C, to 358 MPa at 350° C, and to approximately 515 MPa at 600 and 700° C. It is anticipated that the strength of the composite will decrease above 700° C due to the likelihood that the composite will simply deform without fracturing since the 7740 matrix is extremely ductile above this temperature. From the load versus displacement curves

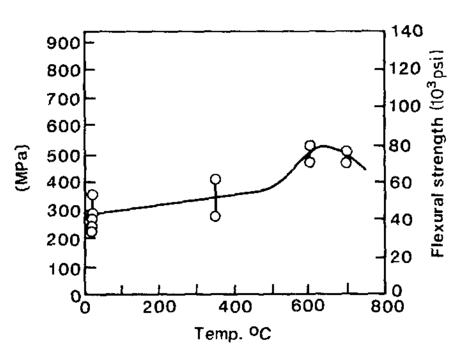

Figure 4 Flexural strength, determined by three-point bend in air, as a function of test temperature for 40 vol% SiC yarn-reinforced 7740 borosilicate glass.

of the SiC yard/glass composites tested in three-point bending, it is apparent that the effective elastic modulus of the composite is decreasing as the temperature is increased with non-linear (plastic) behaviour observed at 700° C. The predominant mode of failure from room temperature to 600° C is local delamination of the composite along the fibre direction, indicative of a weak bond at the fibre matrix interface. Virtually all of the samples tested remained in one piece after testing.

In both the case of the monofilament and yarn-reinforced composites, it is believed that the increase in observed flexural strength with increasing test temperature is due to the fact that the higher test temperature lowers the viscosity and effective yield stress of the matrix. This permits more extensive specimen yielding through the specimen thickness. As a result of this yielding, the actual stress on the specimen tensile surface is relieved and the calculated level of maximum flexural stress can exceed that obtained by the simple beam equation. Thus, the load-carrying capability of the three-point beam has indeed increased with increasing test temperature, however, not necessarily due to an increase in material maximum flexural strength. At the maximum test temperature, general specimen yield occurred at a lower load level as indicated by the calculated decrease in strength.

3.2. Fracture toughness

The fracture toughness of SiC monofilament/7740 composites was measured at 22 and 600° C in the notched three-point bend configuration. Sample dimensions were approximately 0.76 cm wide by 0.56 cm thick by 6.35 cm long. The notch depth was 0.28 cm and the span was 5.08 cm. The load versus deflection curves for the 22 and 600° C tests for 35 vol% SiC composites are shown in Fig. 5. The load to failure increased linearly at both temperatures with the critical stress intensity factors at 22° C of 18.9 MN $m^{-3/2}$ and at 600° C of 14.4 MN $m^{-3/2}$ being calculated from the maximum load prior to failure [7]. The 22° C fracture toughness sample is shown after test in Fig. 6. Zyglo dye penetrant was used to enhance the crack visibility during optical examination. The sample is still in one piece with crack deflection along the fibre direction at the tip of the notch being very apparent. The K_{IC} value of 18.9 MN $m^{-3/2}$ for the SiC-7740 composite is not

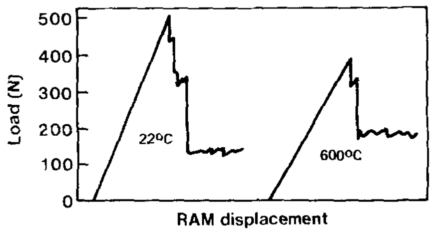

Figure 5 Applied load versus loading ram displacement curves for the testing of pre-notched specimens of 35 vol% SiC monofilament-reinforced 7740 borosilicate glass.

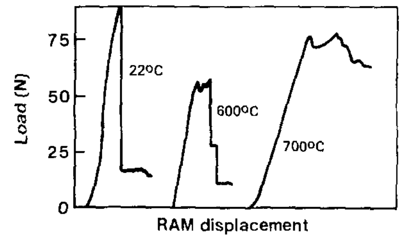

Figure 7 Applied load versus loading ram displacement curves for the testing of pre-notched specimens of 40 vol% SiC yarn-reinforced 7740 borosilicate glass.

significantly different from the K_{IC} at room temperature of 2024-T6 aluminium alloy of 22 MN m$^{-3/2}$.

Notched three-point bend specimens of the SiC yarn/7740 matrix system were also fabricated and tested at room temperature, 600 and 700° C. In this case, specimen dimensions were approximately 0.508 cm thick by 0.254 cm wide by 5.08 cm long. The notch depth was 0.254 cm with a span of 4.00 cm. The load versus deflection curves and stress intensity values determined are shown in Fig. 7. The critical stress intensity factors were calculated in the same manner as for the SiC monofilament/7740 composites. None of the samples failed completely, with crack branching occurring at the notch tip at the lower temperatures. The 700° C sample exhibited no visible crack formation at the tip of the notch even though the sample had undergone considerable deformation. While the K_{IC} values of 11.5 MN m$^{-3/2}$ at 22° C, 7.0 MN m$^{-3/2}$ at 600° C, 8.7 MN m$^{-3/2}$ at 700° C are not as high as those recorded for the SiC monofilament/7740 glass matrix system, they are considerably higher than those of unreinforced glassy materials.

3.3. Composite thermal expansion

The thermal expansion of both monofilament- and yarn-reinforced composites was measured in the axial and transverse directions over the 22 to 550° C temperature range. Composite thermally induced strain was measured as a function of temperature with the average values of coefficient of thermal expansion calculated being presented in Table I. These values were computed using the change in dimension measured between 22 and 500° C and indicate a relatively minor anisotropy in thermal expansion as compared with other composite systems such as graphite-reinforced glass [4].

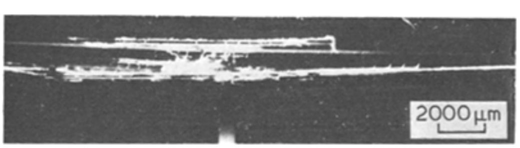

Figure 6 Pre-notched fracture toughness specimen of 35 vol% SiC monofilament-reinforced 7740 borosilicate glass after testing at 22° C. Cracks revealed by use of dye penetrant.

The close agreement in value for the longitudinal and transverse direction for the SiC fibre–7740 matrix composite material is significant in that large anisotropy in thermal expansion coefficients of composites can cause microcracking when the composites are fabricated in cross-plied configurations. No such problems are anticipated for the SiC–7740 system.

3.4. Fibre/matrix interface

During low-temperature flexure testing of the SiC monofilament 7740 composites, it was noticed that failure of the composite invariably occurred through the mechanism of interfacial splitting along the fibre/matrix interface, indicating that the interfacial bonding was very weak. While a weak interfacial bond can be beneficial for fracture toughness, due to crack deflection at the fibre/matrix interface, it can be detrimental to the off-axis properties of the composite. It was thought that the weak bonding could be due to a carbon-rich surface on the SiC fibre. Thus, a series of heat treatments in air was performed in

TABLE I 7740 glass-matrix composite coefficients of thermal expansion (average value between 22 and 500° C)

Filament	Orientation	C.T.E. (10^{-6} ° C)
35 vol% SiC monofilament	0°	4.20
	90°	4.60
40 vol% SiC yarn	0°	3.25
	90°	2.70

TABLE II Properties of SiC fibre-reinforced 7740 glass

	Monofilament		Yarn
Fibre content (vol%)	35	65	40
Density ($g\ cm^{-3}$)	2.6	2.9	2.4
Axial flexural strength (MPa)			
22° C	650	830	290
350° C	–	930	360
600° C	825	1240	520
22° C axial elastic modulus (GPa)	185	290	120
Axial fracture toughness ($MN\ m^{-3/2}$)			
22° C	18.8	–	11.5
600° C	14.3	–	7.0

order to burn off the carbonaceous surface material. It was found that heating the SiC fibre to 800° C or greater in air changed its colour from grey to blue with a corresponding change in the electrical conductivity of the surface of the fibre from conductive to non-conductive. This heat treatment appeared to improve the SiC–7740 bond strength somewhat, but not enough to change the mode of fracture away from being primarily interfacial splitting.

From the fracture characteristic of the SiC yarn glass-matrix system, it appears that the crack blunting ability of this system at its present state of development is somewhat less than that for the SiC monofilament glass system. Although no evidence of a chemical bond between the fibres and matrix can be detected from electron microscopy of fracture surfaces (Fig. 8), fibre pull-out and crack deflection along the fibre/matrix interface is definitely less prevalent for the SiC yarn/glass system than for the SiC monofilament/glass system. This may be a consequence of both the chemical nature of the fibre–matrix bond and the fibre surface to volume ratio. Also,

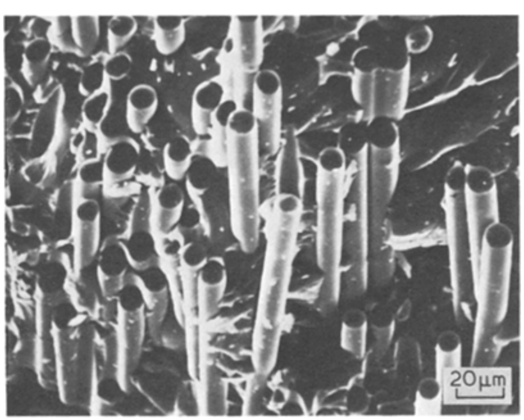

Figure 8 Fracture surface of SiC yarn-reinforced 7740 borosilicate glass.

the strength obtainable for this composite system may be less than that for the SiC monofilament system, owing to the generally lower strength of the SiC yarn. However, owing to the flexibility of the SiC yarn and its ability to be woven into 2-D and 3-D arrays, the potential for fabricating intricate shapes from SiC yarn/glass composites is much greater than for SiC monofilament/glass composites.

4. Conclusions

From the results of this exploration of the SiC fibre-reinforced glass-matrix composite system, it was demonstrated that a material with excellent mechanical properties could be achieved. The physical and mechanical properties of this system are summarized in Table II. With both monofilament- and yarn-type fibres it was possible to achieve high levels of strength and fracture toughness, up to relatively high temperatures. Of the two systems tested, the monofilament-reinforced composite exhibited the higher levels of strength, stiffness and toughness due to the higher properties of the starting fibre. The yarn-reinforced glass composites exhibited, in all cases, less extensive cracking upon fracture due to the much smaller interfibre spacing provided. Although not shown to be important by the data of the current report, this factor may become much more important in fatigue and multiaxial stress state environments.

It can be concluded that the silicon carbide-reinforced glass system can provide flexural strengths equivalent to those obtainable with graphite fibre-reinforced glass and at the same time should be unaffected by exposure to air at elevated temperatures. This enhanced environmental stability has been achieved at the expense of an increase in density of from $2\ g\ cm^{-3}$ for graphite/glass [4] to 2.4 to $2.9\ g\ cm^{-3}$. These values, however, are still less than those for monolithic metal alloys and ceramics which could be applied in this same temperature regime.

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