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Thermal diffusivity and conductivity of a carbon fibre-reinforced borosilicate glass

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A study was conducted of the thermal diffusivity, thermal conductivity and specific heat of hot-pressed Pyrex borosilicate glass reinforced with randomly oriented carbon fibres perpendicular to the hot-pressing direction. The thermal diffusivity and conductivity of the composite perpendicular to the plane of the fibres was only slightly higher than the corresponding value for the matrix, attributed to the low thermal conductivity of the carbon fibres perpendicular to their axis. Within the plane of the fibres the thermal diffusivity and conductivity were approximately twice those of the matrix. The specific heats of the matrix and composite were nearly identical. The temperature dependence of the thermal diffusivity of the matrix and composite was minor only. The corresponding positive temperature dependence of the thermal conductivity of the matrix and composite was attributable primarily to the positive temperature dependence of the specific heat. Heating the composite to temperatures above ~600° C resulted in a permanent decrease in thermal diffusivity at a rate which depended strongly on temperature. This change was attributed to the relaxation of the elastically bent fibres by the thermally activated viscous flow of the surrounding matrix, which as shown by electron microscopy resulted in interfacial separation and crack formation within the composite. These effects, in turn, caused the observed decrease in thermal diffusivity.

1. Introduction

Glass, glass–ceramic and ceramic matrix composites reinforced with fibres or whiskers represent a new class of material for potential applications involving simultaneously high temperatures and high mechanical load. Compared to many single-phase brittle structural materials, these composites exhibit greatly improved fracture toughness and resistance to failure due to mechanical impact and surface damage [1–11]. For service conditions at elevated temperature the thermophysical characteristics of these composites, such as thermal diffusivity, conductivity and specific heat, are critical for purposes of energy efficiency, temperature control, the prevention of failure due to thermal shock [12] and other criteria, governed by the design requirements for any specific structure or component.

The values of the thermophysical properties of the reinforcing fibres or whiskers rarely, if ever, are the same as the corresponding values for the matrix phase. For this reason, composites for any given matrix phase are expected to exhibit thermophysical characteristics which can differ significantly from the values of the matrix, as governed by the chemical composition and structure of the reinforcing fibres or whiskers, their volume fraction, orientation and geometry, and the effect of an interfacial thermal barrier resistance [13–22].

Earlier studies [23, 24] showed that the heat conduction characteristics of high-temperature composites are a function of prior thermal and processing history as well. Composites consisting of a continuous silicon carbide matrix made by the chemical vapour impregnation of amorphous silicon carbide fibres showed a significant permanent increase in the thermal diffusivity on return to ambient temperature following heating to ~1800° C [23]. This effect resulted from the increase in the degree of crystallinity and change in composition of the silicon carbide at temperatures which exceeded temperature at which the fibres were synthesized.

More recently, a uniaxially carbon fibre-reinforced lithia–alumino silicate glass–ceramic was observed to exhibit a permanent decrease in the thermal diffusivity perpendicular to the fibre direction on return to room temperature following heating to temperatures above ~350° C [24]. This effect was thought to be a result of cracks which were observed to form within the matrix. During subsequent thermal cycles the thermal diffusivity perpendicular to the fibre direction exhibited a pronounced hysteresis, which was attributed to the existence of a thermal-history-dependent thermal barrier resistance at the matrix–fibre interface.

It is the purpose of this paper to present observations for the thermal diffusivity of a carbon fibre-reinforced borosilicate glass which also was found

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to exhibit a time-dependent permanent decrease in thermal diffusivity on thermal cycling to high temperatures.

2. Experimental details

2.1. Materials

The matrix phase consisted of a borosilicate glass (Corning Glass Works) with density $\sim 2.23\,\mathrm{g\,cm^{-3}}$ and composition identical to Pyrex glass. Its coefficient of thermal expansion was $\sim 3.6 \times 10^{-6}$ $(^\circ\mathrm{C})^{-1}$.

The carbon fibres consisted of the type known as Celion 6000 (Celanese Corporation), the latter number indicating the number of fibres per tow. Young's modulus of the individual fibres was ~ 235 GPa and the mean fibre diameter $\sim 7\,\mu$m.

The fibres were received in the form of discontinuous fibre mat with a fibre length of ~ 1.9 cm. Plies of this carbon fibre mat were cut to size, impregnated with a slurry containing the glass powder, dried and stacked in a carbon tool and consolidated by hot-pressing at appropriate temperatures and pressures. The resultant composites in the form of a panel measuring 10.2 cm × 10.2 cm × 20.0 cm had a random distribution of the carbon fibres perpendicular to the hot-pressing direction. The coefficient of thermal expansion in the plane of the fibres was $\sim 1.7 \times 10^{-6}$ $(^\circ\mathrm{C})^{-1}$ with a corresponding value perpendicular to the plane of the fibres of $\sim 4.3 \times 10^{-6}$ $(^\circ\mathrm{C})^{-1}$. The mechanical behaviour of these composites was reported previously [25].

Fig. 1 shows an optical micrograph of a polished section perpendicular to the plane of the fibres. The circular or near-circular cross-section of the fibres is indicative of the preferred orientation of the fibres within the hot-pressed disc.

Figs 2a and b show scanning electron micrographs of fracture surfaces parallel and perpendicular to the plane of the fibres. Again, the preferred orientation of the fibres is clearly evident. The generally clean fracture along the interface is indicative of the relatively low degree of adhesion between the carbon fibres and the glass matrix phase.

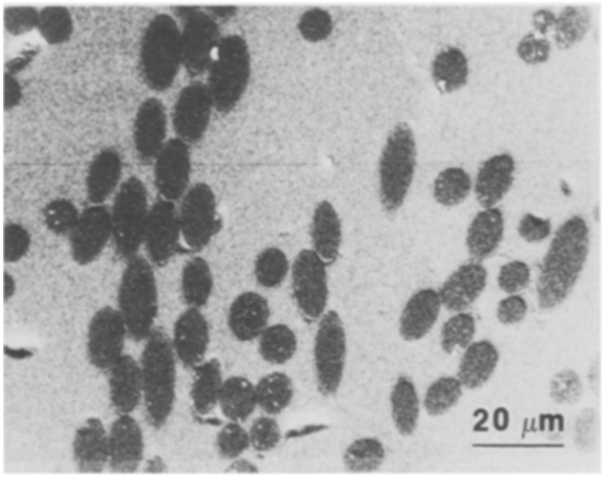

Figure 1 Optical micrograph of polished section of carbon fibre-reinforced borosilicate glass.

2.2. Measurement of thermophysical properties

The thermal diffusivity was measured by the flash method [26] using a neodymium glass laser as the flash source. The specimens were in the form of ~ 8 mm × 10 mm × 2 mm rectangular platelets cut from the hot-pressed billet with a slow-speed precision diamond saw. Specimens were cut to permit measurement of the thermal diffusivity parallel as well as perpendicular to the plane of the fibres. Possible transmission of the laser beam was prevented by coating the specimens with a thin layer of colloidal carbon. The transient temperature response of the specimen rear surface was monitored by remote optical sensing, with a viewing spot size on the specimen ~ 5 mm in diameter. This viewing area is sufficiently large compared to the scale of the microstructure that the detector senses the average temperature of the specimen surface. For this reason, complexities are avoided in evaluating the thermal diffusivity due to local surface temperature differences which arise from differences in the thermal conductivity and diffusivity of the fibres and the matrix [27, 28].

For measurements above room temperature the specimens were held in a carbon resistance furnace with a nitrogen atmosphere. In the evaluation of the thermal diffusivity from the transient temperature response, the effects of the finite laser pulse width and heat losses were taken into account using the analysis of Heckman [29] as well as the changes in the specimen thickness due to thermal expansion. It was found that for temperatures above 600° C the composite specimens exhibited a permanent increase in thickness perpendicular to the fibre plane. This change in thickness could not be monitored while the specimens were at the higher temperature. For this reason, the reported values for the thermal diffusivity for the data obtained on the heating part of the thermal cycle were based on the value of thickness of the specimens prior to heating adjusted for thermal expansion. The dimension of the specimen obtained at room temperature following heating and cooling permitted the calculation of the thermal diffusivity obtained during the cooling part of the cycle. Preliminary observations also indicated that for temperatures above $\sim 600^\circ$ C, the thermal diffusivity exhibited a permanent decrease. In order to investigate this effect the composite samples were held at the highest temperature for a period of ~ 4 h, with data obtained during the heating, holding and cooling parts of the thermal cycle.

The specific heat was determined by differential scanning calorimetry from room temperature to $\sim 550^\circ$ C. The combined data for the thermal diffusivity, specific heat and density were used to calculate the effective thermal conductivity K by means of the relation

$$K = \kappa \varrho c \qquad (1)$$

where κ is the thermal diffusivity, ϱ is the density and c is the specific heat. In the calculation of the thermal conductivity, the decrease in density due to thermal expansion was taken into account.

Because of the permanent changes in specimen

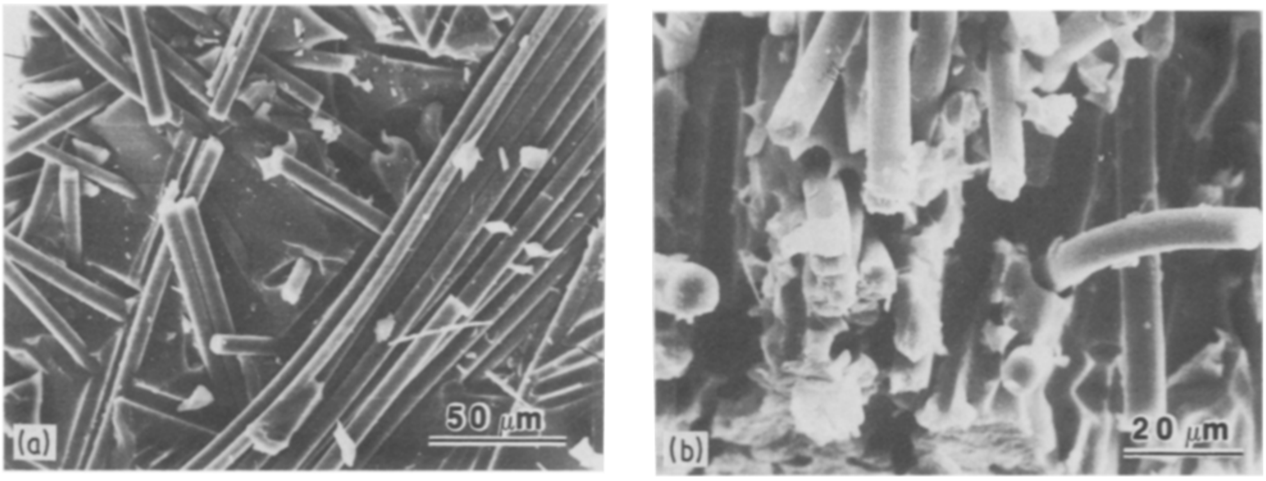

Figure 2 Scanning electron fractographs of carbon fibre-reinforced borosilicate glass; (a) parallel and (b) perpendicular to plane of fibres.

dimensions and thermal diffusivity above 600° C, the experimental data for the thermal diffusivity and specific heat and the calculated values for thermal conductivity for $T < 600$° C will be reported first, followed by the observations for the thermal diffusivity for $T > 600$° C.

3. Results and discussion

3.1. Thermal diffusivity, specific heat and thermal conductivity for temperatures below 600° C

Fig. 3 shows the experimental data for the thermal diffusivity for samples of the glass matrix alone and with the carbon fibre reinforcement for heat flow parallel to the fibre plane. The data for the specific heat are listed in Table I. Fig. 4 shows the values for the thermal conductivity calculated from the smooth curves drawn through the experimental data for the thermal diffusivity shown in Fig. 3, and from the data for the specific heat and the density.

Measurements of the thermal diffusivity of other composite samples at room temperature showed some degree of scatter. For heat flow parallel to the fibre plane, a total of nine samples yielded a mean value of $1.77 \times 10^{-6}\,\mathrm{m^2\,sec^{-1}}$ with a coefficient of variation of 1.9%. Maximum and minimum values were 1.82×10^{-6} and $1.72 \times 10^{-6}\,\mathrm{m^2\,sec^{-1}}$, respectively. Similarly, for heat flow perpendicular to the fibre plane, seven samples exhibited a mean value of $0.855 \times 10^{-6} \pm 3.6\%\,\mathrm{m^2\,sec^{-1}}$, with maximum and minimum values of 0.912×10^{-6} and $0.811 \times 10^{-6}\,\mathrm{m^2\,sec^{-1}}$, respectively. This variation of thermal diffusivity for different composite samples is expected in view of the relatively small specimen size and most likely is attributable to differences in fibre content and/or degree of preferred orientation from sample to sample.

The near temperature-independence of the thermal diffusivity of the glass matrix indicated by the data of Fig. 3 is typical for amorphous dielectric materials with a phonon mean free path of the order of the interatomic distances and not controlled by temperature-dependent phonon collision processes. A similar lack of a pronounced temperature dependence also is exhibited by the carbon fibre-reinforced glass matrix composite. This implies that the thermal diffusivity of the fibres also must be relatively temperature-independent, as the direct result of phonon and electron mean free paths controlled by the high density of lattice defects. A very high density of lattice defects of these fibres is suggested by their very low values of Young's modulus of ~ 235 GPa and thermal

TABLE I Specific heat of soda–alumino–borosilicate glass matrix without and with ~25% carbon fibre reinforcement

Temperature (° C)	Specific Heat ($\mathrm{J\,g^{-1}\,°C^{-1}}$)	
	Glass matrix	Glass with carbon fibre
25	0.767	0.759
50	0.808	0.809
100	0.872	0.898
150	0.927	0.981
200	0.975	1.054
250	1.023	1.107
300	1.066	1.161
350	1.158	1.209
400	1.202	1.243
500	1.255	1.325
550	1.328	1.352

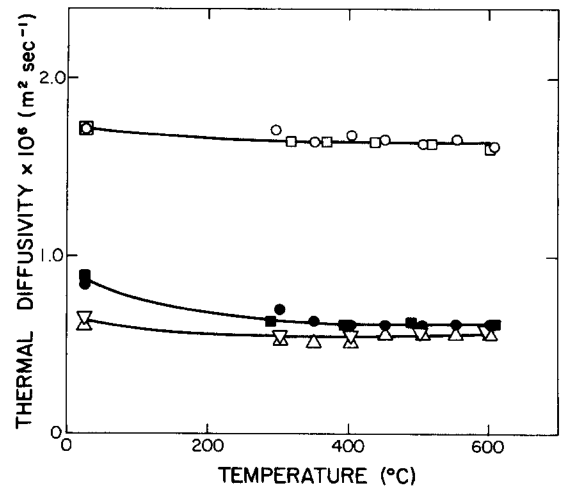

Figure 3 Experimental data for the thermal diffusivity of borosilicate glass matrix with and without carbon fibre reinforcement held at 600° C for 4 h. Parallel to fibre plane (○) heating, (□) cooling; perpendicular to fibre plane (●) heating, (■) cooling; matrix (△) heating, (▽) cooling.

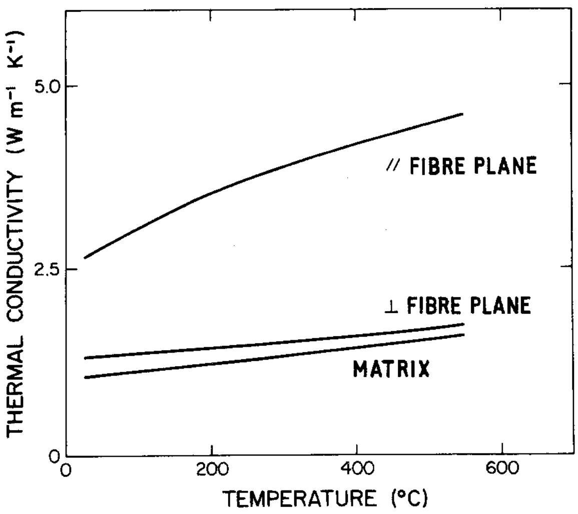

Figure 4 Thermal conductivity of borosilicate glass matrix with and without carbon fibre reinforcement.

conductivity parallel to the fibre axis of $\sim 15\,\mathrm{W\,m^{-1}\,K^{-1}}$ at room temperature, which are much less than the corresponding values for carbon fibres as high as 967 GPa and $\sim 2000\,\mathrm{W\,m^{-1}\,K^{-1}}$, reported by Heremans *et al.* [30].

The coincidence of the data for the thermal diffusivity of the matrix and the composite samples obtained on heating and cooling implies that over the range of room temperature to ~ 600° C, the thermal diffusivity is not affected by irreversible effects due to microstructural or compositional changes.

The data for the specific heat for the matrix phase with and without the carbon fibres are almost identical. This suggests that over the temperature range of this study, the values of specific heat of the fibres and the glass matrix are comparable.

The data for the thermal conductivity in Fig. 4 suggest a number of effects. The near temperature independence of the thermal diffusivity for the matrix as well as composite samples for both directions of heat flow implies that the positive temperature dependence of the thermal conductivity must be attributed primarily to the positive temperature dependence of the specific heat.

The values for the thermal conductivity of the matrix phase of this study up to temperatures of ~ 300° C show excellent agreement with those of Kingery [31], obtained by direct measurement with the comparative method with specimens ~ 2.5 cm thick. Above 300° C, however, the data of Kingery exceed those of the present study by as much as 30% at ~ 600° C. It is thought that this difference at least in part can be attributed to the existence of a partial contribution of radiative heat transfer across the specimen to the total heat conducted. For such radiative heat transfer direcly across the specimen, the effective radiative thermal conductivity increases with increasing specimen thickness [32]. Furthermore, radiative heat transfer is expected to increase with increasing temperature. For these combined reasons, the higher values and corresponding higher temperature dependence of the data of Kingery [31] compared with those for the matrix phase of this study at least in part can be attributed to the differences in specimen thickness.

For the composite, the thermal conductivity perpendicular to the fibre plane is only slightly above the corresponding value for the matrix. This suggests that for this direction of heat flow the fibres have relatively little effect on the thermal conductivity. This is expected for several reasons. Firstly, for this direction of heat flow the matrix phase is continuous and will play a governing role in determining the effective thermal conductivity of the composite. Secondly and possibly more importantly, heat flow within the fibres perpendicular to the fibre axis at least in part will involve heat flow perpendicular to the graphite basal plane. In this direction, as indicated by experimental data for pyrolytic graphite [33–35], the thermal conductivity exhibits rather low values, much lower than those for heat flow within the basal plane. Finally, due to the low degree of adhesion between the fibres and matrix and the mismatch in the coefficients of thermal expansion, the transfer of heat through the composite samples also may be affected by the existence of a thermal barrier resistance at the matrix–fibre interface. Such a thermal barrier resistance, as shown by Hasselman and Johnson [22], generally lowers the effective thermal conductivity of composites.

If it is assumed that such an interfacial thermal barrier resistance is absent, an estimate for the thermal conductivity of the fibres perpendicular to their axes can be obtained from the theory for the effective thermal conductivity for a composite in the form of a matrix with dispersions of non-interacting circular cylinders oriented perpendicular to the direction of heat flow. The expression is derived strictly for parallel circular cylinders. However, as long as the volume fraction is sufficiently dilute, the solution should also apply to the random orientation of fibres within a plane perpendicular to the heat flow as long as the volume fraction is sufficiently dilute that interaction between the local temperature fields around the fibres is absent. Assuming this latter condition is satisfied for the composites of this study, the effective thermal conductivity K_c of the composite perpendicular to the plane of the fibres is [12]

$$K_c = K_m \left[\left(\frac{K_f}{K_m} - 1 \right) V_f + 1 + \frac{K_f}{K_m} \right] \Big/ \left[\left(1 - \frac{K_f}{K_m} \right) V_f + 1 + \frac{K_f}{K_m} \right] \tag{2}$$

where K_m and K_f are the thermal conductivity of the matrix and fibres, respectively and V_f is the volume fraction of fibres.

With $K_c \sim 1.3\,\mathrm{W\,m^{-1}K^{-1}}$, $K_m \sim 1.1\,\mathrm{W\,m^{-1}K^{-1}}$ at room temperature as indicated by the data shown in Fig. 4 and $V_f = 0.25$, Equation 2 yields $K_f \sim 2.2\,\mathrm{W\,m^{-1}K^{-1}}$. This value is in good agreement with the value of $2.4\,\mathrm{W\,m^{-1}K^{-1}}$ for as-deposited unannealed pyrolytic graphite at room temperature reported by Null *et al.* [35]. Of course, caution should be exercised in a direct comparison as the structure of the present fibres with the basal plane more or less circurlarly symmetric around the fibre does not

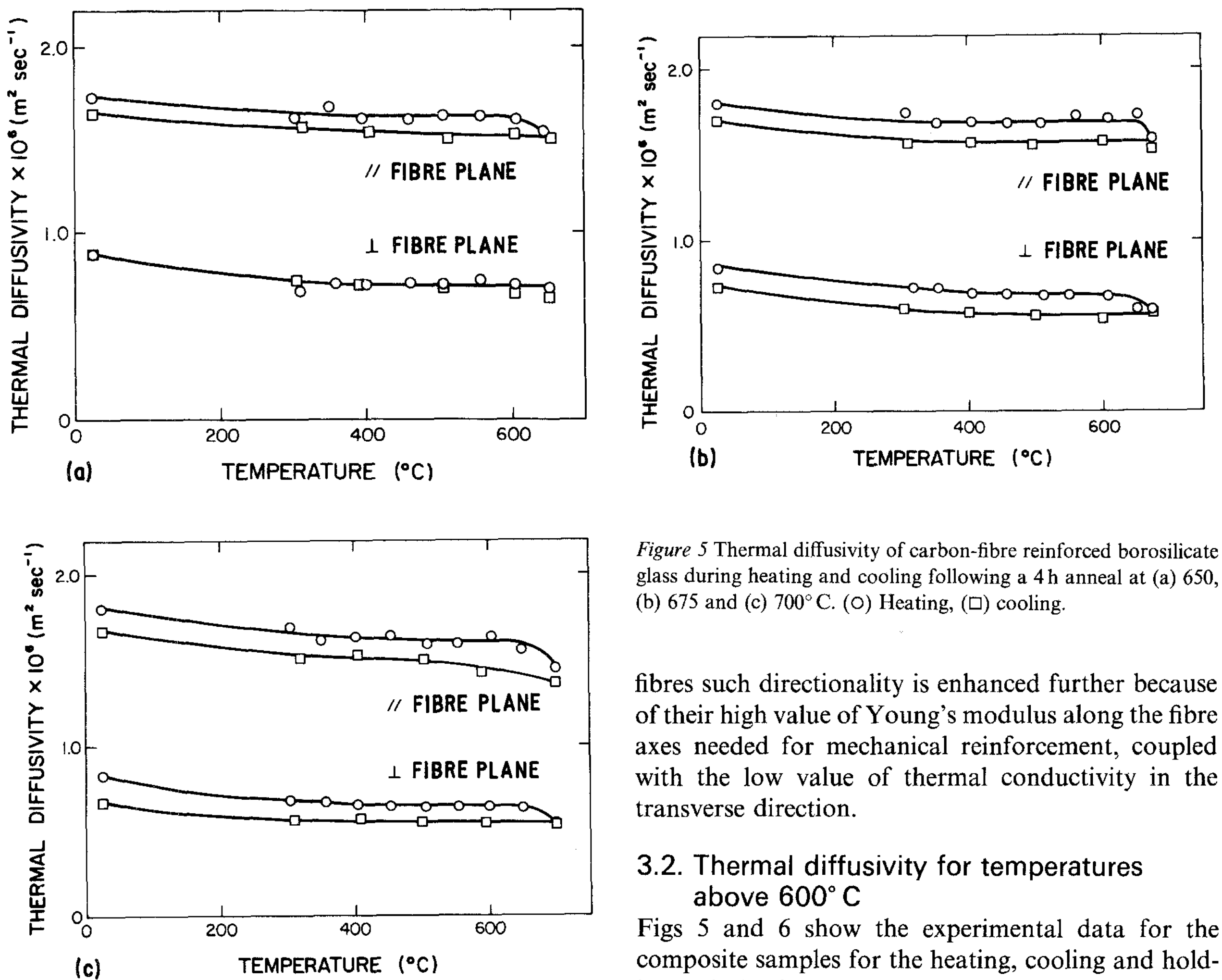

Figure 5 Thermal diffusivity of carbon-fibre reinforced borosilicate glass during heating and cooling following a 4 h anneal at (a) 650, (b) 675 and (c) 700° C. (○) Heating, (□) cooling.

correspond to the planar structure of pyrolytic graphite. Nevertheless, heat flow in the fibres perpendicular to the fibre axis, in part, must occur perpendicular to the basal plane. In this direction, as judged by data for pyrolytic carbon, the thermal conductivity is much lower than within the graphite basal plane [31–33]. For this reason, in terms of a series circuit type of argument, the thermal conductivity perpendicular to the basal plane is expected to play a governing role in establishing the effective thermal conductivity of the fibres perpendicular to the axes. Fibres not oriented perfectly perpendicular to the direction of heat flow, in view of their much higher thermal conductivity along their axis, also could make a contribution to the thermal conductivity perpendicular to the fibre plane. This would involve heat flow at an angle to the temperature gradient, which for a quantitative estimate requires future theoretical analysis. Any contribution of this effect may be offset by the existence of an interfacial thermal barrier resistance. In general, the relatively small increase in thermal conductivity perpendicular to the fibre plane, in contrast to the relatively much larger increase in thermal conductivity parallel to the fibre plane, may well be of practical advantage. In service, a material may have to exhibit high mechanical load-bearing ability in one direction and be a good thermal insulator in a transverse direction. Generally, composite materials such as those of this study exhibit such directional properties for fibre orientations other than fully random. For carbon fibres such directionality is enhanced further because of their high value of Young's modulus along the fibre axes needed for mechanical reinforcement, coupled with the low value of thermal conductivity in the transverse direction.

3.2. Thermal diffusivity for temperatures above 600° C

Figs 5 and 6 show the experimental data for the composite samples for the heating, cooling and holding periods of thermal cycles to ~ 650, 675 and 700° C. In order to more effectively indicate the relative effect of temperature, the data in Fig. 6 were plotted on a normalized basis, which eliminates the effect of the sample-to-sample variation in the thermal diffusivity prior to heating, referred to earlier. Included in Fig. 6 are the data for the samples heated to 600° C, for which data obtained during heating and cooling are presented in Fig. 3. Table II lists the data for the thickness of the samples at room temperature prior to and following completion of the thermal cycle.

The data of Fig. 5 for cycling to all three temperatures indicate a permanent decrease on return to

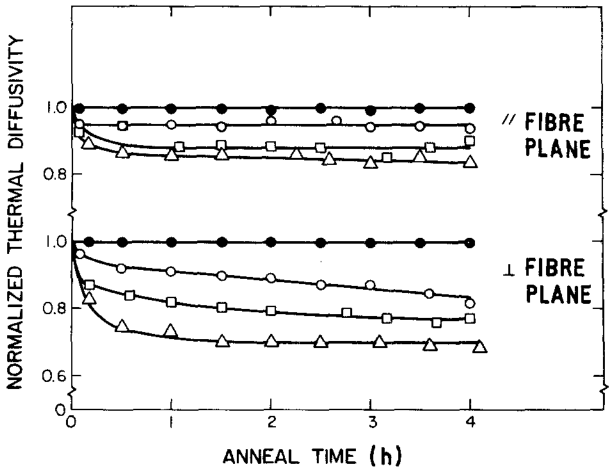

Figure 6 Relative thermal diffusivity of carbon fibre-reinforced borosilicate glass during 4 h anneal at (●) 600, (○) 650, (□) 675 and (△) 700° C for thermal cycles shown in Figs 3 and 5.

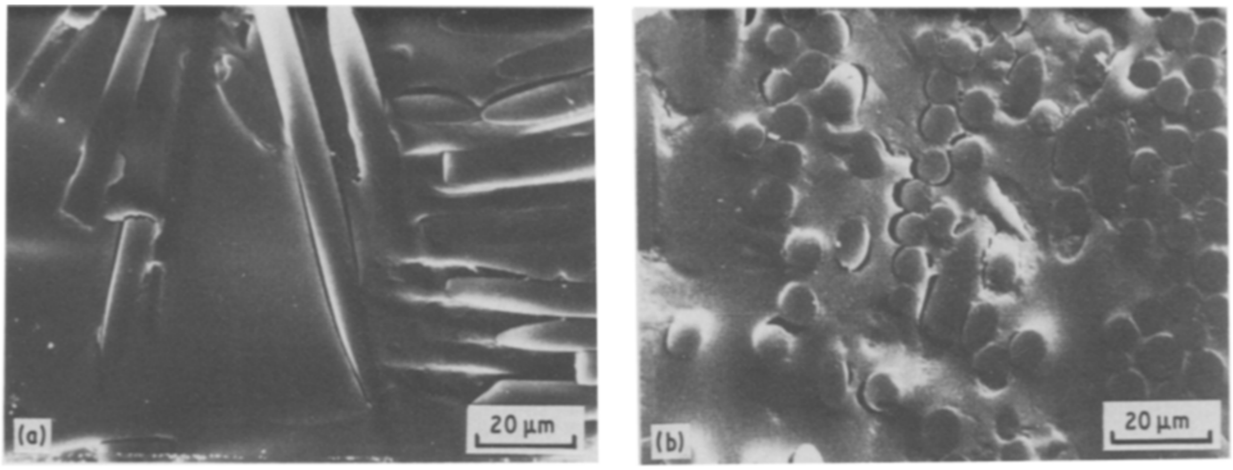

Figure 7 Scanning electron micrographs of sections of carbon fibre-reinforced borosilicate glass annealed at 650° C for 4 h following polishing: (a) parallel, (b) perpendicular to fibre plane.

room temperature, indicative of a permanent structural change. The increase in dimensions of the specimens, which is particularly pronounced perpendicular to the fibre plane, suggests that this structural change takes the form of the formation of voids or cavities. The data of Fig. 6 suggest that the mechanism responsible for such void or cavity formation is governed by a thermally activated process. The data of Fig. 6 also show that the thermal diffusivity decreases asymptotically to a constant value. This suggests that cavity formation is of a transient nature only.

The scanning electron micrographs shown in Fig. 7 of polished surfaces perpendicular and parallel to the fibre plane show that the cavity formation took the form of interfacial cracking between the fibres and the matrix. Such interfacial cracking also appeared to involve displacement of the fibres, which appeared to protrude above the surface as shown by the micrograph of the polished section parallel to the fibre plane in Fig. 7b.

It is speculated here that the above permanent structural change originates from the elastic bending of the fibres over one another by the compaction during hot-pressing. Because the compacts were cooled while under pressure, over the lower ranges of temperatures the fibres are held in place by the rigid matrix. However, on re-heating the composites without external constraints up to a temperature sufficiently high for the matrix to exhibit viscoelastic behaviour, stress relaxation of the fibres can occur by the thermally activated stress-enhanced viscous flow of the glass phase. The viscosity data presented by Shand [36] for Pyrex glass suggest temperatures for the softening, annealing and strain points of 821, 560 and 510° C, respectively. These data suggest that above 600° C the matrix phase should show significant viscous deformation. The associated cavity formation by crack formation or interfacial separation lowers the thermal diffusivity as the direct result of the presence of the cracks [37] and/or increase in the thermal barrier resistance at the fibre–matrix interface [21].

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References

1. K. M. PREWO and J. J. BRENNAN, *J. Mater. Sci.* **15** (1980) 463.
2. *Idem, ibid.* **17** (1982) 1201.
3. J. J. BRENNAN and K. M. PREWO, *ibid.* **17** (1982) 2371.
4. T. MAH, M. G. MENDIRATTA, A. P. KATZ, R. RUH and K. S. MAZDIYASNI, *J. Amer. Ceram. Soc.* **68** (1985) C-27.
5. F. P. MEYER, G. D. QUINN and J. C. WALCK, *Ceram. Eng. Sci. Proc.* **6** (1985) 646.
6. R. RICE, *ibid.* **6** (1985) 589.

TABLE II Dimensional changes of carbon fibre-reinforced Pyrex glass

Maximum temperature of cycle (° C)	Heat flow with respect to fibre plane	Thickness (cm)	
		Original	After heating
600	∥	0.190	0.190
	⊥	0.195	0.196
650	∥	0.203	0.203
	⊥	0.191	0.203
675	∥	0.190	0.191
	⊥	0.195	0.208
700	∥	0.191	0.191
	⊥	0.194	0.212

7. E. FITZER and R. GADOW, *Amer. Ceram. Soc. Bull.* **65** (1986) 326.
8. P. J. LAMICQ, G. A. BERNHART, M. M. DAUCHIER and J. G. MACE, *ibid.* **65** (1986) 336.
9. D. P. STINTON, A. J. CAPUTO and R. A. LOWDEN, *ibid.* **65** (1986) 347.
10. P. D. SHALEK, J. J. PETROVIC, G. F. HURLEY and F. D. GAC, *ibid.* **65** (1986) 351.
11. K. P. GADKAREE and K. CHYUNG, *ibid.* **65** (1986) 370.
12. D. P. H. HASSELMAN, *J. Amer. Ceram. Soc.* **52** (1969) 600.
13. Lord RAYLEIGH, *Phil. Mag.* **34** (1892) 481.
14. J. C. MAXWELL, “A Treatise on Electricity and Magnetism”, 3rd Edn (Oxford University Press, 1904).
15. H. FRICKE, *Phys. Rev.* **24** (1924) 575.
16. D. A. G. BRUGGEMAN, *Ann Phys.* **24** (1935) 636.
17. E. H. KERNER, *Proc. Phys. Soc. (London)* **B69** (1956) 802.
18. Z. HASHIN, *J. Compos. Mater.* **2** (1968) 284.
19. S. C. CHENG and R. I. VACHON, *Int. J. Heat Mass Transfer* **12** (1969) 249.
20. B. BUDIANSKY, *J. Compos. Mater.* **4** (1970) 286.
21. S. NOMURA and T. W. CHOU, *ibid.* **14** (1980) 120.
22. D. P. H. HASSELMAN, L. F. JOHNSON, *ibid.* (in press).
23. H. TAWIL, L. D. BENTSEN, S. BASKARAN and D. P. H. HASSELMAN, *J. Mater. Sci.* **20** (1985) 3201.
24. D. P. H. HASSELMAN, L. F. JOHNSON, R. SYED, M. P. TAYLOR and K. CHYUNG, *ibid.* **22** (1987) 701.
25. K. M. PREWO, *ibid.* **17** (1982) 3549.
26. W. J. PARKER, R. J. JENKINS, C. P. BUTLER and G. L. ABBOTT, *J. Appl. Phys.* **32** (1961) 1679.
27. M. S. DESHPANDE, R. H. BOGAARD and R. E. TAYLOR, *Int. J. Thermophys.* **2** (1981) 357.
28. R. E. TAYLOR, *High Temp.–High Press.* **15** (1983) 299.
29. R. C. HECKMAN, *J. Appl. Phys.* **44** (1973) 1455.
30. J. HEREMANS, C. P. BEETZ Jr, I. RAHIM and M. S. DRESSELHAUS, Proceedings of Conference “Thermal Conductivity 19” edited by D. Yarbrough (Plenum, New York).
31. W. D. KINGERY, *J. Amer. Ceram. Soc.* **42** (1959) 617.
32. W. D. KINGERY, H. K. BOWEN and D. R. UHLMANN, “Introduction to Ceramics”, 2nd Edn. (Wiley, New York, 1976).
33. S. NASU, T. TAKAHASHI and T. KIKUCHI, *J. Nucl. Mater.* **43** (1972) 72.
34. T. TANAKA and H. SUSUKI, *Carbon* **10** (1972) 253.
35. M. R. NULL, W. W. LOZIER and A. W. MOORE, *ibid.* **11** (1973) 81.
36. E. B. SHAND, “Glass Engineering Handbook”, 3rd Edn (McGraw-Hill, New York, 1984).
37. D. P. H. HASSELMAN, *J. Compos. Mater.* **12** (1978) 403.

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