JOURNAL OF MATERIALS SCIENCE 17 (1982) 2337–2342

Determination of the thermal conductivity and diffusivity of thin fibres by the composite method

J. J. BRENNAN
United Technologies Research Center, East-Hartford, CT 06108, USA

L. D. BENTSEN, D. P. H. HASSELMAN
Department of Materials Engineering, Virginia Polytechnic Institute and State University, Blacksburg, VA 24061, USA

It is suggested that the thermal conductivity of very fine fibres can be evaluated indirectly with the aid of composite theory using the experimental data for the heat transport properties of an appropriate composite which contains the fibres. The feasibility of this approach was investigated by determining the thermal conductivity and diffusivity of fibres of amorphous silicon carbide from 25° C to 1000° C contained within a lithium aluminosilicate glass-ceramic using the laser-flash technique for measurement of the thermal diffusivity of the composite. Due to the amorphous nature of the fibres, values for their thermal conductivity and diffusivity were found to be far less than the corresponding data for crystalline silicon carbide. The positive temperature dependence of the thermal conductivity, coupled with the independent observation of an increase in thermal conductivity with specimen thickness, suggests that radiative heat transfer makes a significant contribution to the total heat transferred. A number of advantages and limitations of the composite method for the evaluation of thermal transport properties of fibres are discussed.

1. Introduction

The technical and scientific need for quantitative data for the heat conduction properties of solids has led to the development of a nearly innumerable number of methods for the measurement of these properties [1]. For a given material, the choice of the most appropriate method depends on a number of variables such as the temperature range over which the data are required and the configuration of the material to be measured. In this respect, micrometre sized fibres present special difficulties.

Materials in fibre form are used extensively in the development of mechanically reinforced composites. One such composite developed during the last few years consists of a glass or glass-ceramic reinforced with fibres of silicon carbide [2]. The mechanical properties of these composites indicate excellent service performance at elevated temperature. For a micro-mechanical analysis of these composites under conditions of transient heat flow, experimental data for the thermal conductivity and diffusivity were required as a function of temperature. The choice of the method for the measurement of these properties presented a number of difficulties. Firstly, their method of preparation precluded the synthesis of larger samples with properties identical to those of the fibres. For this reason, the fibres themselves had to be used. Secondly, silicon carbide fibres are not electrical conductors. This eliminated the resistive heating method [3] for measuring the thermal conductivity. Although, in principle, a number of techniques could be devised, it was decided to measure the thermal conductivity and diffusivity of these fibres by an indirect method. This method

0022–2461/82/082337–06$03.22/0

consisted of measuring the thermal diffusivity of the composite as well as the matrix (without fibres), followed by calculation of the required data by means of the theory for the thermal conductivity of composites. It is the purpose of this paper to report the data so obtained.

2. Experimental procedure

2.1. Materials

The silicon carbide fibres made from an organometallic polymer by the method of Yajima [4] were obtained from a commercial source*. Chemical analysis showed the fibres to consist of approximately 65 wt% SiC, 25 wt% SiO_2 and about 10 wt% C. The fibres were in the form of tows of yarn containing about 500 fibres per tow. The average fibre diameter was approximately 10 μm and the density was approximately 2.55 g cm^{-3}. X-ray analysis showed the fibres to be amorphous with a crystallite size of 2.5 to 3.0 nm, determined from peak broadening.

The matrix material with density of 2.52 g cm^{-3} consisted of a lithium aluminosilicate glass-ceramic essentially identical to that of a commercial glass-ceramic† except that the approximately 3 wt% TiO_2 nucleating agent was replaced by about 2 wt% ZrO_2 due to reactivity between the TiO_2 and the SiC fibres.

The composite was manufactured by passing the SiC yarn through a slurry of powder of the uncrystallized glass and isopropyl alcohol. Following drying, the coated yarn was cut to fit the graphite die for hot-pressing in vacuum for times of between 5 min and 1 h at 1400 to 1500° C and a pressure of 14 MPa. Following hot-pressing, crystallization of the glass was carried out by heat-treating for 1 to 2 h at a temperature ranging from 880 to 1100° C. Composite samples were made in which the fibres were all parallel or with alternate layers oriented at 90°. Fig. 1 shows a photomicrograph of such a 0/90° cross-ply composite. A sample of the matrix phase without fibres was also prepared.

2.2. Measurements of the thermal properties

The thermal diffusivity of the composite and matrix samples was measured by the laser-flash method [5] using a Nd-glass laser. The specimens used for these measurements were cut from the hot-pressed blanks, and were in the form of square plates of dimensions 9 to 12 mm × ~ 2 mm. Direct transmission of the laser beam was prevented by coating the specimen surfaces with carbon. The transient temperature response of the specimen rear surface was monitored by optical means. For measurements above room temperature the specimens were held in an appropriate holder contained within a carbon resistance furnace with nitrogen atmosphere. In the evaluation of the thermal diffusivity from the transient temperature response, the corrections for heat-loss were taken into account using the analysis given by Heckman [6]. Changes in specimen thick-

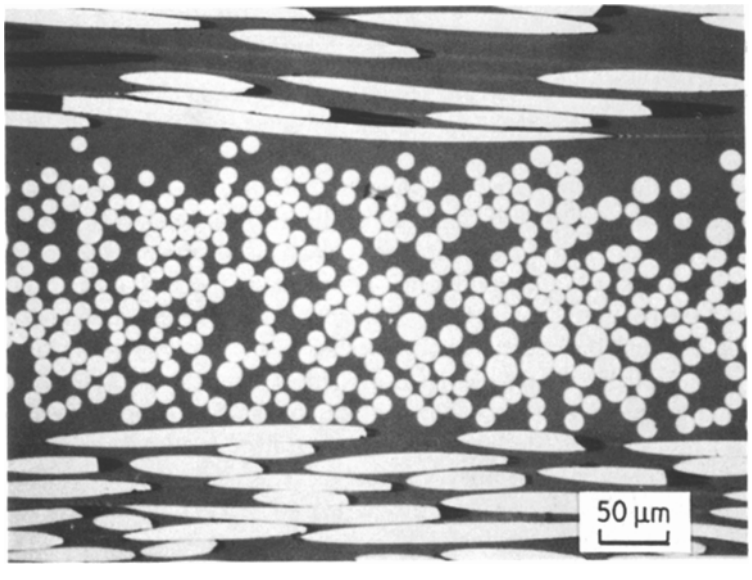

Figure 1 Optical micrograph of 0/90° composite of lithium aluminosilicate glass-ceramic with 45 vol% amorphous silicon carbide fibres.

*Nippon Carbon Company, Japan.
†C-9608, Corning Glass Works, USA.

ness with temperature due to thermal expansion were also taken into account.

The specific heat of the fibres and the glass-ceramic matrix was determined by differential scanning calorimetry using appropriate equipment‡.

2.3. Evaluation of the thermal conductivity and diffusivity of the fibres from composite theory

For heat-flow parallel to uniaxially aligned fibres, the thermal conductivity, K_c, of a composite is

$$K_c = K_m V_m + K_p V_p, \quad (1)$$

where K is the thermal conductivity, V is the volume-fraction and the subscripts c, m and p, refer to the composite, matrix and fibres, respectively.

For heat-flow perpendicular to the fibre direction, the thermal conductivity, as derived by Bruggeman [7], can be written

$$\left(\frac{K_m - K_c}{K_m + K_c}\right) V_m = \left(\frac{K_c - K_p}{K_c + K_p}\right) V_p. \quad (2)$$

From the measured value of the thermal diffusivity, the corresponding value of the thermal conductivity, K, can be calculated from

$$K = \kappa \rho c, \quad (3)$$

where κ is the thermal diffusivity, ρ is density and c is the specific heat. The specific heat of the composite can be calculated from the measured values for the specific heat of the fibres and the matrix by means of the rule of mixtures. Substitution of the values for the thermal conductivity of the matrix and the composite into Equation 1 or 2 permits calculations of the thermal conductivity of the fibres. The thermal diffusivity may then be determined using Equation 3.

3. Experimental results

Fig. 2 shows the experimental data for the specific heat as a function of temperature for the glass-ceramic matrix and the silicon carbide fibres. The data for the glass-ceramic agree very well with those obtained from the manufacture of a glass-ceramic of similar composition [8]. The specific heat of the silicon carbide fibres significantly exceeds the corresponding values for crystalline silicon carbide [9]. The difference can most probably be attributed to the non-crystalline nature of the fibres, since large relative differences in the specific heat of amorphous and crystalline polymers are also observed [10].

Table I lists the experimental data for the thermal diffusivity at room temperature for a number of composite samples of different thicknesses with a range of volume-fraction and both fibre orientations. Also included in Table I are the values for the room temperature thermal conductivity and thermal diffusivity of the fibres calculated from the data for the specific heat, density and thermal diffusivity.

Fig. 3 shows the experimental data for the temperature dependence of the thermal diffusivity of the glass-ceramic matrix and two composite

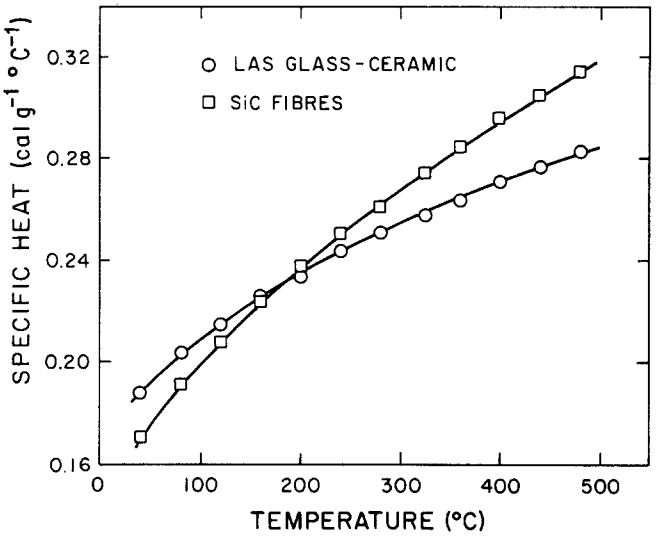

Figure 2 Specific heat of lithium aluminosilicate glass-ceramic and of amorphous silicon carbide fibres.

‡Dupont 990 thermal analyser.

TABLE I Thermal diffusivity at room temperature of LAS-SiC composites with a range of sample thicknesses and SiC contents and calculated values for the thermal diffusivities and conductivities of the SiC fibres

Measured values for composites				Calculated values for SiC fibres	
SiC content (vol%)	Orientation of fibres with respect to the direction of heat-flow	Thickness (mm)	Thermal diffusivity ($\times 10^{-5}$ cm^2 sec^{-1})	Thermal diffusivity ($\times 10^{-5}$ cm^2 sec^{-1})	Thermal conductivity (W m^{-1} K^{-1})
48	⊥	1.020	722	642	1.10
48	⊥	1.520	824	845	1.44
48	⊥	2.250	837	873	1.49
48	∥	1.050	744	669	1.14
48	∥	1.540	818	831	1.42
48	∥	2.130	828	854	1.46
48	∥	2.690	826	848	1.45
48	∥	2.785	849	899	1.53
45	⊥	0.975	735	653	1.12
45	⊥	1.485	806	803	1.37
45	⊥	2.275	855	915	1.56
38	⊥	1.235	710	587	1.17
38	⊥	1.245	753	685	1.00
32	⊥	1.305	739	624	1.07

samples containing ~ 49 vol% SiC with heat-flow parallel and perpendicular to the fibres. The data for the LAS matrix show reasonable agreement with data obtained earlier [11] for a sample of a similar glass-ceramic (C9608) supplied by the manufacturer.

Fig. 4 shows the values for the thermal conductivity and thermal diffusivity of the silicon carbide fibres parallel and perpendicular to the fibre axis, calculated from the experimental data given in Figs 2 and 3.

4. Discussion

The values of the thermal conductivity and thermal diffusivity of the silicon carbide fibres are far below the corresponding values of crystalline silicon carbide [12, 13]. The low values for the present fibres are most likely the result of their amorphous nature which limits the contribution of the phonon conductivity.

The relatively small negative temperature dependence of the thermal diffusivity and the positive temperature dependence of the thermal conductivity of the fibres is an indication that a significant fraction of the total heat conducted occurs by radiative heat transfer. Additional independent support for the latter conclusion is also provided by the observations, shown in Table I, that the thermal diffusivity of the composite samples with the SiC fibres increases with increasing specimen thickness, in accordance with theoretical expectations [14]. Assuming that at room temperature the radiation contribution is negligible, the data shown in Fig. 4 suggest that at

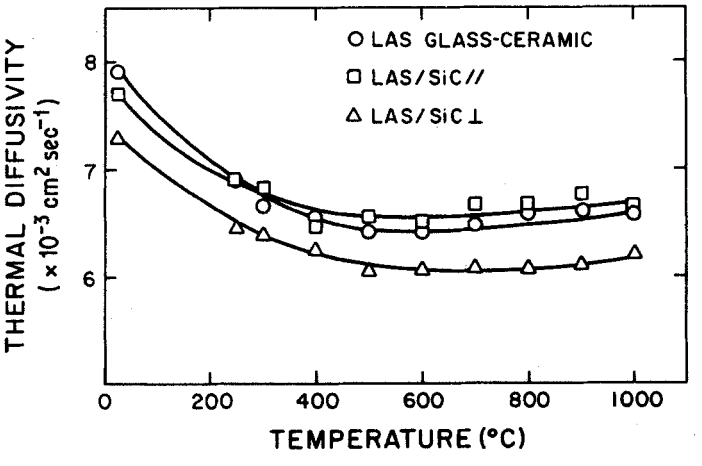

Figure 3 Experimental data for the thermal diffusivity as a function of temperature for lithium aluminosilicate glass-ceramic matrix with 0, 48 and 49 vol% SiC fibres for specimen thicknesses of 1.32, 1.29 and 1.29 mm, parallel and perpendicular to the fibre axis, respectively.

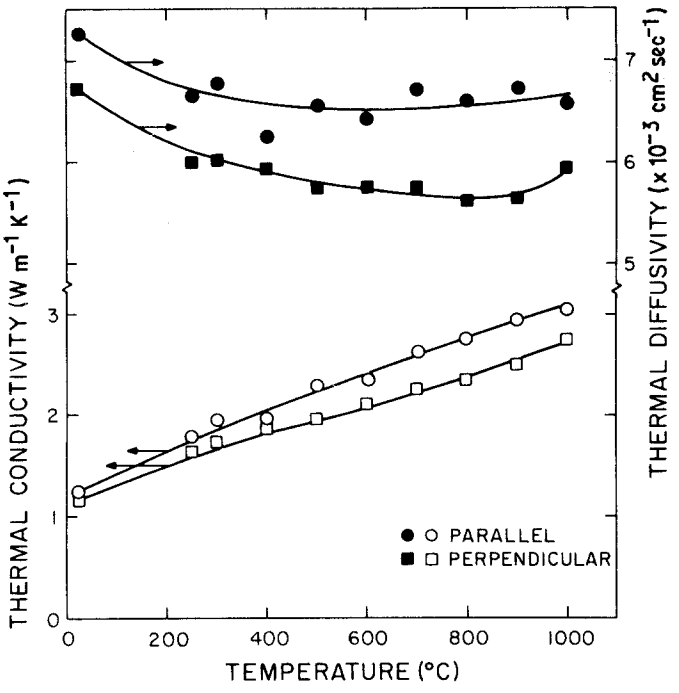

Figure 4 Temperature dependence of the thermal conductivity and diffusivity of amorphous silicon carbide fibres calculated from the composite data shown in Fig. 3.

1000° C the radiation contribution is in excess of half of the total heat transferred. This suggests that, although no supporting experimental data appear to be available, these SiC fibres are possibly excellent transmitters of infra-red radiation of relatively long wavelength.

The thermal diffusivity and conductivity perpendicular to the fibre axis is somewhat less than parallel to the fibre axis. This could possibly be attributed in part to a structural anisotropy within the fibre resulting from its original organic structure. No such preferred orientation, however, was detected by Yajima *et al.* [4]. Possibly also, an interfacial contact resistance due to a lack of perfect adhesion or the factor of 2 to 3 difference in the thermal expansion between the glass-ceramic matrix and the fibres could be responsible for this effect. This would manifest itself as an apparent decrease of the thermal conductivity perpendicular to the fibre axis, because Equation 3 does not take such an interfacial resistance into account. Such an interfacial resistance would have a much smaller effect (if any) on heat-flow parallel to the fibre axis. For this reason the evaluation of the fibre conductivity by the composite method should preferably rely on experimental data using uniaxially aligned composite samples with heat-flow parallel to the fibre length.

Some general remarks are in order on the general feasibility of the "composite method" for obtaining heat conduction data for fibres. It is critical to note that this method relies on the accuracy of the theory for the thermal conductivity of composites. In general, such theory is exact only for dilute concentrations of the second phase. At higher volume-fractions, at which the local temperature fields around the disperions will interact, the composite theory must be considered to be an approximation only. Any deviations between composite values calculated from such approximations and actual values increase with the relative difference in the values of the thermal conductivity of the individual phases. For this reason, increased accuracy of the value for the thermal conductivity of fibres obtained by the "composite method" can be achieved by choosing a matrix material with a value of the thermal conductivity as close as possible to that of the fibres. This condition is met for the composites of this study. For the same reason, the choice of the composite equation used to calculate the thermal conductivity of the SiC fibres is not critical.

Furthermore, for the "composite method" to yield reliable data for the heat transport properties of the fibres, it is essential that the scale of the microstructure and the size of the sample of the composite is such that, in its transient response, the sample, in effect, behaves as a continuum. Criteria for this condition were examined by Kerrisk [15, 16], Lee and Taylor [17], and Nomura and

Chou [18]. For the samples used in the present study, these criteria were met.

In summary, it is proposed that the thermal conductivity of fibres can be obtained from the corresponding data for composites by means of the theory for the thermal conductivity of composites. The data presented indicate the feasibility of this method.

Acknowledgement

The present study was supported by the Office of Naval Research under contract numbers N00014-78-C-0431 and N00014-78-C-0503.

References

1. Y. S. TOULOUKIAN, R. W. POWELL, C. Y. HO and P. G. KLEMENS, "Thermal Conductivity – Nonmetallic Solids" (IFI/Plenum Press, New York and Washington, DC, 1970) pp. 13a–25a.
2. K. M. PREWO and J. J. BRENNAN, *J. Mater. Sci.* **15** (1980) 463.
3. Y. S. TOULOUKIAN, R. W. POWELL, C. Y. HO and P. G. KLEMENS, "Thermal Conductivity – Nonmetallic Solids" (IFI/Plenum Press, New York and Washington, DC, 1970) pp. 19a–22a.
4. S. YAJIMA, K. OKAMURA, J. HAYASHI and M. OMORI, *J. Amer. Ceram. Soc.* **59** (1976) 324.
5. W. J. PARKER, R. J. JENKINS, C. P. BUTLER and G. L. ABBOTT, *J. Appl. Phys.* **32** (1961) 1679.
6. R. C. HECKMAN, *ibid.* **44** (1973) 1455.
7. D. A. G. BRUGGEMAN, *Annal. Physik* **24** (1935) 636.
8. "Materials Handbook", Corning Glass Works, 1961.
9. Y. S. TOULOUKIAN and E. H. BUYCO, "Specific Heat – Non-metallic Solids" (IFI/Plenum Press, New York and Washington, DC, 1970) pp. 448–450.
10. R. A. V. RAFF and K. W. DOAK (EDS), "Crystalline Olefin Polymers, Part II" (John Wiley and Sons, New York, 1964) p. 216.
11. D. P. H. HASSELMAN, unpublished data (1973).
12. Y. S. TOULOUKIAN, R. W. POWELL, C. Y. HO and P. G. KLEMENS, "Thermal Conductivity – Nonmetallic Solids" (IFI/Plenum Press, New York and Washington, DC, 1979) pp. 585–588.
13. Y. S. TOULOUKIAN, R. W. POWELL, C. Y. HO and M. C. NICOLAOU, "Thermal Diffusivity" (IFI/Plenum Press, New York and Washington, DC, 1973) pp. 447–478.
14. W. D. KINGERY, H. K. BOWEN and D. R. UHLMANN, "Introduction to Ceramics", 2nd edn. (John Wiley and Sons, New York, 1976) pp. 627–634.
15. J. F. KERRISK, *J. Appl. Phys.* **42** (1971) 267.
16. *Idem, ibid.* **43** (1972) 112.
17. T. Y. R. LEE and R. E. TAYLOR, *J. Heat Trans.* **100** (1978) 720.
18. S. NOMURA and T. W. CHOU, *J. Composite Mater.* **14** (1980) 120.

Received 30 November
and accepted 15 December 1981