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The transverse coefficient of thermal expansion of a unidirectional composite

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Various analytical models of the effective thermal expansion coefficients of unidirectional fibre-reinforced composite materials predict for certain fibre–matrix combinations an increase in the transverse coefficient of thermal expansion over that of its constituents at low fibre volume content. This effect is especially noticeable if the composite is fabricated with fibres of high modulus and low thermal expansion coefficient in matrices of low modulus and high thermal expansion coefficient. An experimental investigation was therefore conducted to study this behaviour in Textron fibre (SCS-6)-reinforced Hercules 3501-6 epoxy matrix. Numerical calculations for this material system have shown that increases of the order of 20% over the matrix expansion coefficient is possible for fibre volume fraction in the range 3%–4%. Experimental measurements of the effective thermal expansion coefficients are seen to be in favourable agreement with the theoretical predictions. A parametric study is also undertaken to examine the influence of constituent properties on the effective composite behaviour. It is shown that the axial restraint of the fibre is responsible for a peak in the behaviour of the transverse expansion coefficient.

1. Introduction

The design of a fibrous composite material to yield the properties desired for a particular application requires an analytical description of the effective properties of the composite based on the geometric arrangement and mechanical properties of its constituents. Consideration of thermoelastic problems for composites requires the definition of composite moduli, thermal conductivities, specific heat and thermal expansion coefficients. A comprehensive review of composite thermoelastic properties is included in Hashin [1].

Evaluation of the response of composite materials to temperature changes is important not only for high- and low-temperature applications but also for fabrication considerations, such as the choice of cure temperature. Thermal expansion behaviour can also be important when composite materials are used in conjunction with other materials and when it is necessary to match the thermal expansion coefficient of one structural component with another for dimensional stability and mechanical compatibility. From an engineering standpoint, the understanding of thermal expansion coefficients of unidirectional composites has become significant because of the wide use of fibrous composites in various applications in recent years. Thermal residual stresses in laminates cannot be calculated without full information about the thermal expansion behaviour and elastic response of unidirectional composites.

In the present paper attention is focused on the effective thermal expansion coefficients of a two-phase unidirectional fibrous composite. Theoretical predictions [2–11] of the effective coefficient of thermal expansion have indicated that the transverse coefficient of thermal expansion of the composite can be higher than that of its constituents at low fibre volume fractions. Some limited experimental measurements of the transverse coefficient of thermal expansion have been made by Schneider [12], Yates *et al.* [13] and Bowles and Tompkins [14], amongst others, at moderate to high fibre volume fractions ($\sim$ 0.3–0.8). However, there is little experimental data for thermal expansion coefficient at low fibre volume fractions.

The purpose of this work was to test the applicability of theoretical models, which predict an interesting phenomenon wherein the composite thermal expansion coefficient is larger than either one of its constituents, by experimentally measuring the transverse thermal expansion behaviour of unidirectional fibre-reinforced composites at low fibre volume fractions (< 0.1). Unidirectional specimens with controlled fibre volume fractions were made and the effective thermal expansion measured. The experimental values were than correlated with the theoretical predictions using a concentric cylinder model [15]. An attempt has also been made to provide a physical explanation for the increase in transverse coefficient of thermal expansion in terms of the internal stress state within the composite. Finally, parametric studies have been conducted to examine the influence of constituent properties on composite behaviour.

2. Theoretical background

The effective thermal expansion coefficients are defined as the average strains resulting from a unit temperature rise for a traction-free material. Effective thermal expansion coefficients of anisotropic composites having any number of anisotropic phases have been bounded from above and below using thermoelastic energy principles by Rosen and Hashin [16]. For isotropic phases, the thermal expansion results reduce to the bounds obtained by Schapery [2]. When the composite has only two phases, thermal expansion coefficients coincide to give exact solutions. The two-phase results for thermal expansion coefficients can be obtained more directly following the methods of Levin [17]. For the unidirectional fibrous composite (one fibre direction) of two isotropic phases, there are two different expansion coefficients (the axial, α_{11}, and the transverse, α_{22}) given by

$$\alpha_{11} = (c_f\alpha_f + c_m\alpha_m) + \left[(\alpha_f - \alpha_m)\bigg/\left(\frac{1}{K_f} - \frac{1}{K_m}\right)\right] \times \left[\frac{3(1-2\nu_{12})}{E_{11}} - \left(\frac{c_f}{K_f} + \frac{c_m}{K_m}\right)\right] \tag{1a}$$

$$\alpha_{22} = \{c_f\alpha_f + c_m\alpha_m\} + \left[(\alpha_f - \alpha_m)\bigg/\left(\frac{1}{K_f} - \frac{1}{K_m}\right)\right] \times \left[\frac{3}{2K_{23}} - \frac{3\nu_{12}(1-2\nu_{12})}{E_{11}} - \left(\frac{c_f}{K_f} + \frac{c_m}{K_m}\right)\right] \tag{1b}$$

where E_{11}, K_{23}, and ν_{12} are the composite effective uniaxial modulus, effective plain strain bulk modulus, and effective axial Poisson's ratio, respectively; c, the phase volume fraction, α, the coefficient of thermal expansion, K, the bulk modulus and the subscripts f and m refer to the fibre and matrix, respectively.

Interestingly, the two-phase effective coefficients of thermal expansion do not require the introduction of a specific geometric model. They depend only on the symmetry characteristics of the equivalent homogeneous medium. However, they do have a dependence on the effective elastic properties, which must be known either from experimental data or from the theoretical prediction based on a particular geometric model. Thus, when the solutions are available for the elastic moduli, the problem is solved for the two-phase composite.

Theoretical predictions of the effective coefficient of thermal expansion by Rosen [3] using a concentric cylinder model have indicated that the transverse coefficient of thermal expansion of the composite can be higher than that of its constituents at low fibre volume fraction. Similar observations have been made by Schapery [2] and Chamis and Sendeckyj [4] amongst other researchers. This effect is especially noticeable with fibres of high modulus and low axial expansion coefficient (e.g. boron or carbon) in a low-modulus matrix having a high coefficient of thermal expansion (e.g. epoxy resin) [5]. The coefficients of thermal expansion have also been computed by Ishikawa *et al.* [6] based on a hexagonal and square array model and have reported observing a maximum in the transverse expansion coefficient for both carbon and glass fibre-reinforced epoxy composites. Schneider's [7] treatment of hexagonal arrangement of fibres accounting for the matrix wedges between the elements similarly reported a maximum in the transverse thermal expansion coefficient of the glass–epoxy system.

Using an infinite series solution based upon the known local elastic field solution derived by the perturbation expansion of the Green's tensor function, Nomura and Chou [8] have derived the effective thermal expansion coefficients for multi-phase short-fibre composites and have numerically shown that a maximum of transverse expansion coefficient occurs at low fibre volume fraction for continuous glass fibre-reinforced epoxy composites. The effective thermal expansion coefficients of an aligned short-fibre composite have also been investigated by Takao and Taya [9, 10] using Eshelby's equivalent inclusion method and have shown that a peak occurs at fibre volume fraction approximately equal to 0.1 in the carbon/epoxy system for large fibre aspect ratios (i.e. continuous fibres).

The influence of internal stresses, due to the thermo-mechanical mismatch between the fibre and the matrix, on the thermal expansion behaviour of unidirectional fibre-reinforced ceramics has recently been considered by Hsueh and Becher [11]. Using a composite cylinder model, they have calculated the total strains (hence, the effective thermal expansion coefficient) directly from the internal thermal stresses and shown numerically that the transverse thermal expansion coefficient of the composite can be higher than that of its constituents.

As mentioned earlier, though various analytical models have indicated an apparent increase in the transverse coefficient of thermal expansion at low fibre volume fraction, no attempts, to our knowledge, seem to have been made to validate this interesting phenomenon by experiment.

3. Experimental procedure

Unidirectional specimens were fabricated using H-3501-6 epoxy resin and CVD silicon carbide fibres (SCS-6). Selected properties of the fibre and the matrix are shown in Table I. This material system was considered as an excellent choice for the test of the theoretical predictions because of the high elastic modulus and low thermal expansion coefficient of the fibre and low modulus and high thermal expansion coefficient of the epoxy matrix. Monofilaments rather than yarns were chosen based primarily on the ability to control the spacing, alignment and volume fraction of the fibres. In order to accomplish these requirements, specially designed aluminium jigs were machined with closely spaced holes arranged in a square pattern. Several of these jigs were made so as to be able to vary the volume fraction of fibres from 1%–8%. The fibres were cut to 50 mm lengths and cleaned in acetone thoroughly to remove all the sizing. The fibres were then inserted in the jig (Fig. 1) and the

TABLE I Selected thermo-mechanical properties of the fibre and matrix

Material	E (GPa)	ν	α ($10^{-6}\,°C^{-1}$)
SCS-6 (fibre)	413.7	0.24	2.4
H 3501-6 (matrix)	4.27	0.34	51.15

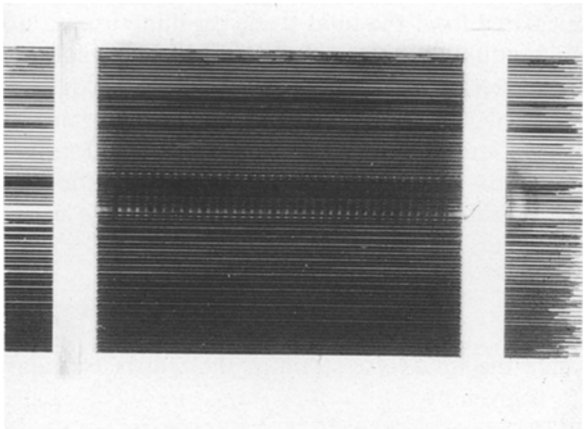

Figure 1 A micrograph showing the fibres in the jig before matrix infiltration.

loaded jig was put in a silicone rubber mould which had already been outgassed so as to eliminate porosity in the resulting composites.

The epoxy resin was liquefied at 120 °C and vacuum debulked for 30 min. The molten resin was transferred into the mould containing the fibres and cured at 120 °C for 30 min and subsequently at 180 °C for 2 h before being slowly cooled back to room temperature. A typical cooling cycle would take several hours. This was done to avoid excessive thermal shock and matrix cracking. Excess matrix was pored into the mould so as to have specimens containing only the matrix made from the same batch of the resin as the actual composites. For every pouring the thermal expansion coefficient of the matrix was measured together with that of the corresponding composites. This was done to ensure that the comparisons between the matrix and the composite properties were made on specimens cured in an identical fashion.

The sample was then taken out of the rubber mould and the aluminium jig was cut off using a high-speed water-cooled diamond saw. The excess matrix sticking to the jig was burnt out at 375 °C for 45 min so that the jig could be reused again. Fig. 2a and b show photographs of a composite specimen after matrix infiltration. Then specimens measuring 12 mm × 4 mm × 5 mm were cut from this composite block for the measurement of the transverse thermal properties. To obtain the highest sensitivity from the measurements, the specimens had its longest dimension transverse to the fibre direction. The specimens containing just the matrix had the same dimensions as the composites. All specimens were cut using a slow speed diamond saw and polished successively up to 600 grit to have flat parallel faces and also to remove any other defects.

The transverse thermal expansion coefficients were then measured using a Du Pont thermomechanical analyser model 9900 interfaced to a computer for data collection and analysis. The machine was calibrated using a tungsten NBS standard before use. From the softening point data on the matrix all thermal expansion measurements were made to temperatures of 175 °C. All tests were done in air. The heating cycle was programmed so as to have soaks at regular intervals of 25 °C up to the maximum temperature. This was done to ensure that the sample had sufficient time at the temperature of interest to equilibrate. The heating rate was chosen as 2 °C min^{-1}. The thermal expansion coefficients were calculated by the computer using a point to point line instead of a best fit between the selected temperatures. Because the composites were transparent, the volume fraction of the fibres were measured by counting the number of monofilaments in each specimen and multiplying it with the cross-sectional area of each fibre and dividing that by the cross-sectional area of the composite.

4. Results and discussion

The effective coefficient of thermal expansion (ECTE) in the transverse direction has been plotted in Fig. 3 as

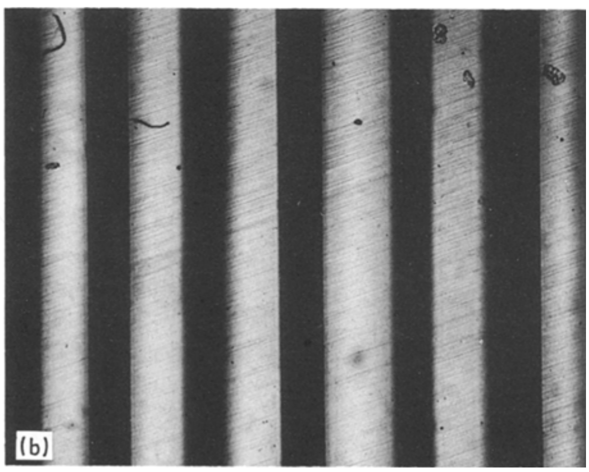

Figure 2 A composite specimen with perfectly aligned fibres after matrix infiltration, (a) cross-sectional view, and (b) top view.

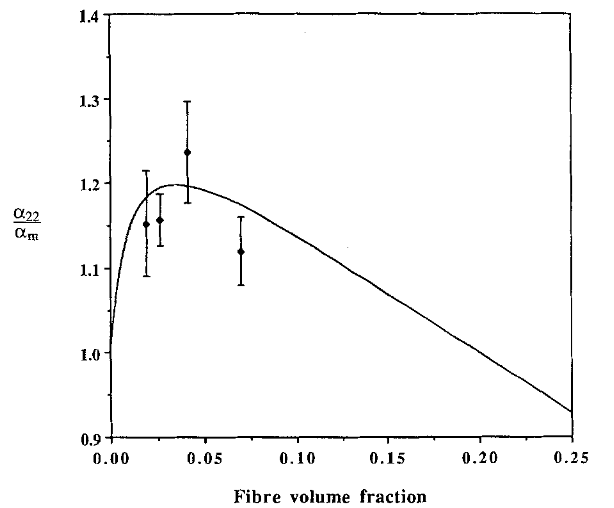

Figure 3 Transverse coefficient of thermal expansion of unidirectional SCS-6/H3501-6 composite. (——) Theory, (♦) experiment.

a function of fibre volume content (the theoretical curve is shown as a solid line and the data points are the experimental values). All the values have been normalized with respect to the matrix expansion coefficient in order to eliminate the variability in the measured values from batch to batch due to processing. Although the fibres were arranged in a square array in the specimens, theoretical results were computed using a concentric cylinder model [15] because few differences are expected between the values calculated from the two models. Moreover, our aim was to demonstrate that the measured property (ECTE) can become larger than that of its constituents, rather than fit the experimental data to a particular theoretical model. It is seen from Fig. 3 that at low fibre volume fraction the transverse coefficient of thermal expansion of the composite initially increases beyond that of the matrix value, peaks at a fibre volume approximately equal to 0.035 and finally decreases with further increase in fibre volume. As is evident from the plot, there seems to be good correlation between the data and the computed values at the various fibre volume fractions investigated, though some measured values were lower and some higher than the theoretical predictions. Nevertheless, all the experimental measurements of α_{22} are certainly greater than that of the matrix at the various fibre volume fractions investigated and do capture the existence of a peak in the transverse thermal expansion coefficient with increasing fibre volume fraction.

As pointed out by Schapery [2], the initial increase in the transverse coefficient of thermal expansion is apparently due to the axial restraint of the fibres. In the longitudinal direction the composite properties are predominately fibre dominated. Because the fibre has a low coefficient of thermal expansion and a high elastic modulus, the matrix in the composite is constrained from expanding in the longitudinal direction even though it (the matrix) has a high coefficient of thermal expansion. However, in the transverse direction the matrix is relatively free to expand. Therefore the longitudinal constraint felt by the matrix is compensated by larger expansion in the transverse direction. With increasing fibre volume fraction, the fibre properties eventually begin to dictate the expansion behaviour of the composite. Because the fibre has a low coefficient of thermal expansion, the overall composite property decreases.

Instead of relating the effective coefficient of thermal expansion to the effective elastic constants (Levin' approach), the ECTE can also be calculated directly from the internal thermal stresses [11]. In that model [11], the effective thermal expansion coefficients were calculated from the total strain resulting from a unit temperature change in the absence of externally applied stresses. The total composite strain, in turn, consisted of mechanical strains due to the internal thermal stress and the unconstrained thermal strain of the matrix. For a composite cylinder model, the transverse coefficient of thermal expansion (α_{22}) can be defined as

$$\alpha_{22} = \frac{\varepsilon_t^m}{\Delta T} \tag{2}$$

where the transverse strain at the matrix boundary, ε_t^m, is given by

$$\varepsilon_t^m = \frac{1}{E_m}(\sigma_t^m - \nu_m \sigma_z^m) + \alpha_m \Delta T \tag{3}$$

and E_m and ν_m denote the matrix Young's modulus and Poisson's ratio, respectively, and σ_t^m and σ_z^m are the transverse and longitudinal stress components, respectively, at the matrix boundary. Hence, if the mechanical strain $[(\sigma_t^m - \nu_m \sigma_z^m)/E_m]$ has the same sign as the unconstrained thermal strain of the matrix ($\alpha_m \Delta T$), the composite coefficient of thermal expansion (α_{22}) will be higher than the thermal expansion coefficient of the matrix (α_m). The distribution of the mechanical strain at the matrix boundary is plotted in Fig. 4 as a function of fibre volume content. As seen from the figure, the strain distribution exhibits a peak at a fibre volume fraction approximately equal to 0.035. Thus, the stress state in the system is seen to induce additional elastic expansion during heating in the transverse direction at the outer surface of the

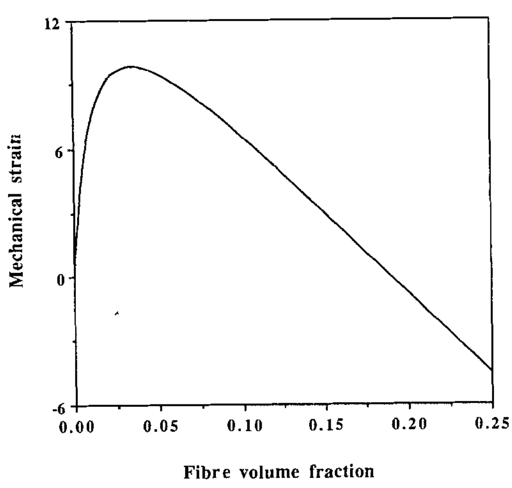

Figure 4 The distribution of the mechanical component of the total strain at the outside radial boundary for a unit temperature change (using a concentric cylinder model [15]).

matrix, and which, in turn, results in a higher coefficient of thermal expansion.

A unidirectional fibre-reinforced material can also be viewed (approximately) as a laminated system consisting of transversely isotropic plies (representative of constituent materials). An exact solution for thickness expansion coefficients of composite laminates has been developed by Pagano [18]. Using the laminate analogy, the expansion coefficient in the thickness direction for a $[0/0]_s$ laminate was computed and a trend similar to that obtained from the composite cylinder assemblage model was observed.

Finally, to place into proper perspective the influence of constituent properties on the effective composite behaviour, we will present some numerical results of the parametric study that was undertaken. Fig. 5 illustrates the variation of the normalized effective thermal expansion coefficient with fibre volume fraction. The ratio of fibre to matrix Young's modulus, E_f/E_m, and matrix to fibre coefficient of thermal expansion, α_m/α_f, were the two variables selected for this study. The matrix and fibre Poisson's ratio were kept constant at 0.34 and 0.24, respectively. For a stiffer matrix ($E_f/E_m < 1$), the effective transverse coefficient of thermal expansion is seen to obey almost a linear relationship with fibre volume fraction for various ratios of the thermal expansion coefficients (Fig. 5a). However, for $E_f > E_m$, which is typical for most fibre-reinforced materials, the effective expansion coefficient is seen to exhibit a maximum for a high value of α_m/α_f and a minimum for a low value of α_m/α_f as seen in Fig. 5b. The influence of the stiffening ratio, E_f/E_m, for $\alpha_m/\alpha_f < 1$ and $\alpha_m/\alpha_f > 1$, is further illustrated in Fig. 5c and d, respectively. When the coefficient of thermal expansion of the matrix is larger than that of the fibre, the composite transverse coefficient of thermal expansion is higher than either one of its constituents for a stiff fibre (large value of E_f/E_m). Conversely, when the fibre has a thermal expansion coefficient larger than the matrix, the composite property is lower than either the fibre or the matrix. It is apparent from the results displayed that it is the axial restraint of the fibre (high ratio of E_f/E_m) which is responsible for a peak in the behaviour of the transverse coefficient of thermal expansion of the composite at low fibre volume fraction. Looking at Fig. 5c and d, it is further observed that when $\alpha_m/\alpha_f < 1$, $E_f = E_m$ condition results in an upper bound of the ECTE, whereas when $\alpha_m/\alpha_f > 1$, $E_f = E_m$ leads to a lower bound of the transverse coefficient of thermal ex-

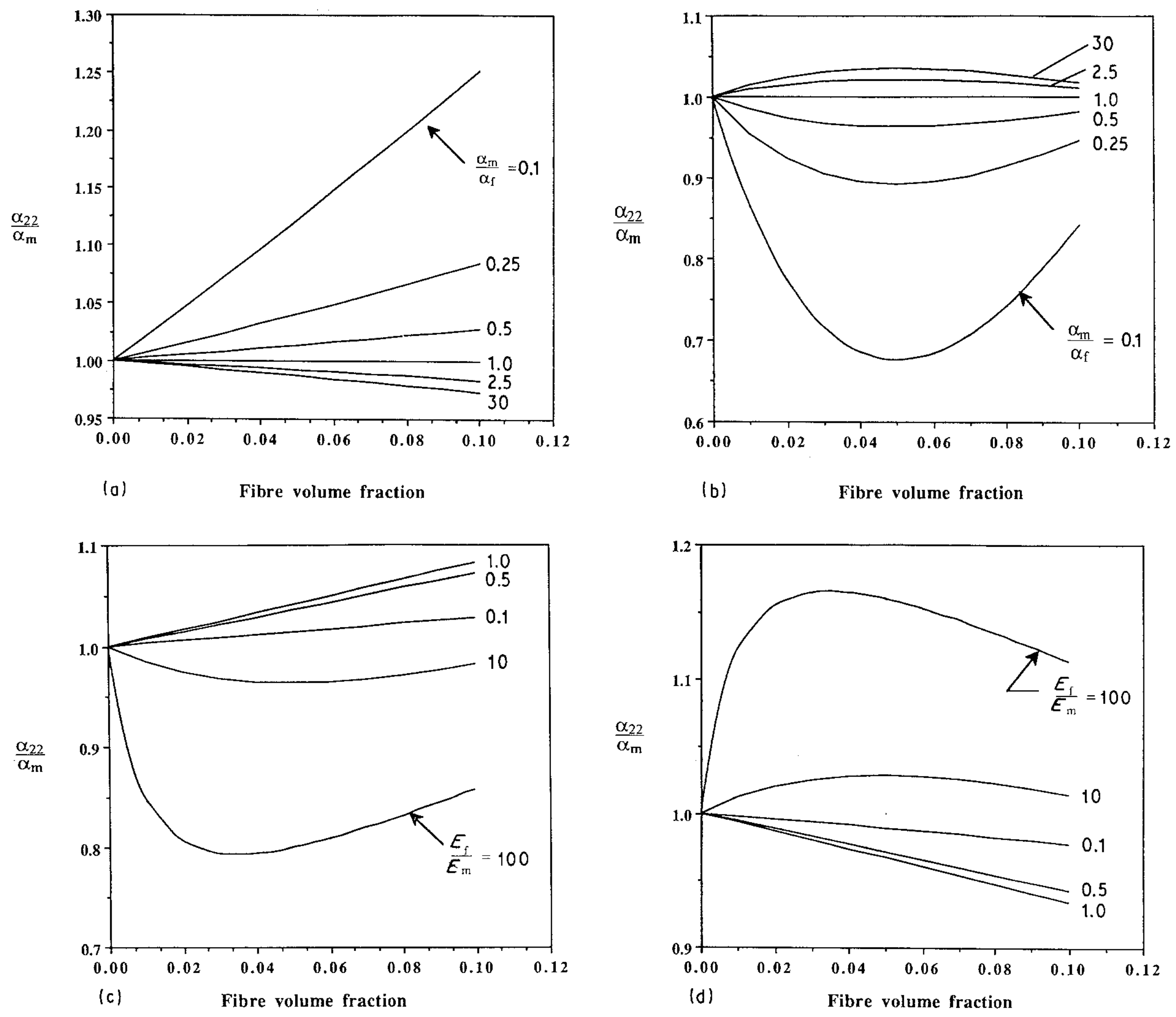

Figure 5 The variation of effective transverse thermal expansion coefficient with fibre volume fraction, V_f, for (a) $E_f/E_m = 0.1$; (b) $E_f/E_m = 10.0$; (c) $\alpha_m/\alpha_f = 0.5$; and (d) $\alpha_m/\alpha_f = 5.0$.

pansion. This observation of the bounding behaviour is being investigated further and will be reported elsewhere.

5. Conclusion
Theoretical predictions of the coefficients of thermal expansion of unidirectional fibre-reinforced composite materials show that for certain fibre–matrix combinations, at low fibre volume fractions, the transverse coefficient of thermal expansion of the composite can be higher than that of its constituents. An experimental investigation was carried out to study this anomalous behaviour in Textron fibre (SCS-6)-reinforced Hercules 3501-6 epoxy matrix. The measured values of the transverse coefficient of thermal expansion are shown to agree very well with the numerical calculations and are certainly greater than that of the matrix at the various fibre volume fractions investigated. The experimental results capture the existence of a peak in the transverse thermal expansion coefficient with increasing fibre volume fraction. It is shown that the axial restraint of the fibre is responsible for a peak in the behaviour of the transverse expansion coefficient.

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References
1. Z. HASHIN, NASA CR-1974 (1972).
2. R. A. SCHAPERY, *J. Compos. Mater.* **2** (1968) 380.
3. B. W. ROSEN, *Proc. Roy. Soc. Lond. Ser. A* **319** (1970) 79.
4. C. C. CHAMIS and G. P. SENDECKYJ, *J. Compos. Mater.* **2** (1968) 332.
5. D. K. HALE, *J. Mater. Sci.* **11** (1976) 2105.
6. T. ISHIKAWA, K. KOYAMA and S. KOBAYASHI, *J. Compos. Mater.* **12** (1978) 153.
7. W. SCHNEIDER, *Kunstoffe* **61** (1971) 273 (in German).
8. S. NOMURA and T. W. CHOU, *Int. J. Engng Sci.* **19** (1981) 1.
9. Y. TAKAYO and M. TAYA, *ASME J. Appl. Mech.* **52** (1985) 806.
10. *Idem., J. Compos. Mater.* **21** (1987) 140.
11. C. HSUEH and P. F. BECHER, *J. Amer Ceram. Soc.* **71** (1988) C-438.
12. W. SCHNEIDER, *Kunststoffe* **63** (1973) 929 (in German).
13. B. YATES, M. J. OVERY, J. P. SARGENT, B. A. McCALLA, D. M. KINGSTON-LEE, L. N. PHILIPS and K. F. ROGERS, *J. Mater. Sci.* **13** (1978) 433.
14. D. E. BOWLES and S. S. TOMPKINS, *J. Compos. Mater.* **23** (1989) 370.
15. N. J. PAGANO and G. P. TANDON, *Compos. Sci. Technol.* **31** (1988) 273.
16. B. W. ROSEN and Z. HASHIN, *Int. J. Engng Sci.* **8** (1970) 157.
17. V. M. LEVIN, *Mech. Solids* **2** (1967) 58.
18. N. J. PAGANO, *J. Compos. Mater.* **8** (1974) 310.

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