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Interfacial Bonding and Thermal Expansion of Fibre-Reinforced Composites

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The present work examines the effect that the fibre-matrix bond strength has on longitudinal and transverse thermal expansivities of unidirectional glass fibre and carbon fibre-reinforced epoxies. It is shown that the weakening of the interfacial bond results in an increase of the longitudinal coefficient of thermal expansion, and a decrease of the transverse one. The results are explained in terms of the reinforcement efficiency factor, according to a theory proposed elsewhere.⁴

INTRODUCTION

Thermal expansion of fibre-reinforced composites depends on various factors, such as the properties of the constituent materials or the geometry of the reinforcement. Being a dilatation strain, it also depends on the interfacial shear transfer mechanism, which determines the extent of axial restraint of the fibres. This restraint is effective in both longitudinal and transverse directions. In the longitudinal direction low expansions are obtained due to the fibres constraining the matrix from expanding, and also, this constraint results in "squeezing" the matrix out in the transverse direction.¹ The interfacial shear transfer mechanism is affected by two factors: the fibre-matrix bond strength, and their contact area. Thus, a reduction in the fibre-matrix bond strength is anticipated to result in an increase of the longitudinal expansivity, with a simultaneous decrease of the transverse expansivity.

The present study was destined to investigate the effect of the fibre-matrix bond strength on the thermal expansivities of unidirectional composite materials. In the course of the study the interfacial bond strength was varied by exposing the test specimens to boiling water for various lengths of time, the water gradually destroying the fibre-matrix bond. The work was carried out with glass fibre and carbon fibre-reinforced epoxies.

EXPERIMENTAL

A plate of glass fibre (Vetrotex, EC14-300-K937)—epoxy resin (Ciba-Geigy Ltd., Araldite MY750/HT972) composite was manufactured as in Ref. 2. Carbon fibre (Courtaulds Ltd., Grafil HT-S)—epoxy resin (Union Carbide, ERLA4617/DDM) composite plate was manufactured from the commercial "pre-pregs". From these plates longitudinal and transverse specimens presented in Figure 1 were cut.

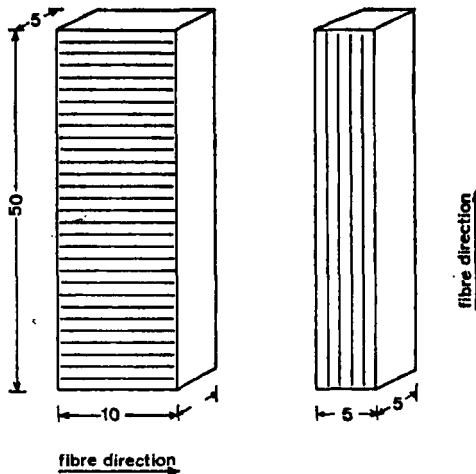


FIGURE 1 Geometries of test specimens used for measuring longitudinal and transverse thermal expansivities. (All measurements are in millimeters.)

The water-boil process was carried out as follows: A test specimen was boiled in water for a length of time, then dried at 110°C for 4 hours; thereafter the thermal expansivity was measured. This cycle was repeated until the boiling period totalled at least 24 hours.

The coefficients of the thermal expansion were measured over the range room temperature to 110°C. The measurements were carried out by means of a stainless steel apparatus described by Wilson and Treloar.³ This

apparatus was calibrated by means of an electrolytic copper specimen of known expansivity ($\alpha = 1.962 \times 10^{-5} \text{ }^\circ\text{C}^{-1}$). The temperature was raised at an average rate of $0.4^\circ\text{C}/\text{min.}$, and readings were taken every 30 minutes. The resulting expansion versus temperature curves were linear,⁴ except at the first point (room temperature), being the only one measured at a steady state. A value of the thermal expansivity was calculated from the slope of each line worked out by a linear regression analysis, excluding the first point. The estimated error was within $\pm 0.004 \times 10^{-5} \text{ }^\circ\text{C}^{-1}$.

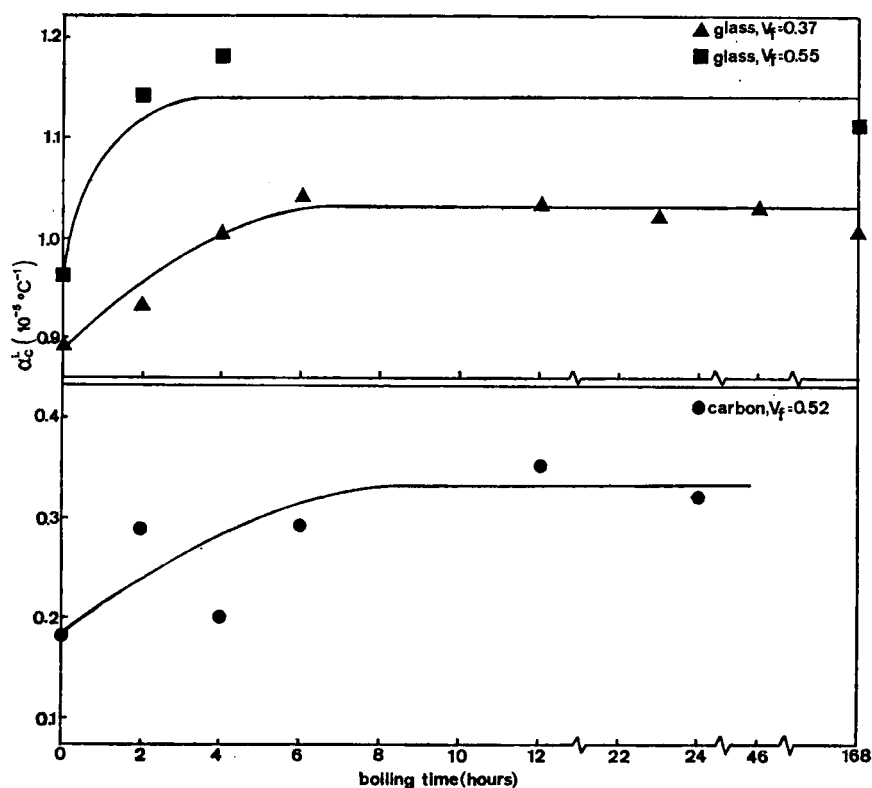


FIGURE 2 Results of longitudinal coefficients of thermal expansion as a function of boiling period.

3. RESULTS

The effect of the water-boil treatment on the longitudinal thermal expansivity (α_L) of glass fibre-reinforced epoxy and of carbon fibre-reinforced epoxy is demonstrated in Figure 2. Similarly, Figure 3 demonstrates this effect on

the transverse thermal expansivity (α_c^T) of these materials. It is seen that both types of composites in either transverse or longitudinal measurements are affected by the exposure to boiling water. This effect is similar for the two types of composites, which reach ultimate expansivities during ~ 5 hours of water-boil treatment. Thereafter the thermal expansivities are independent of this treatment.

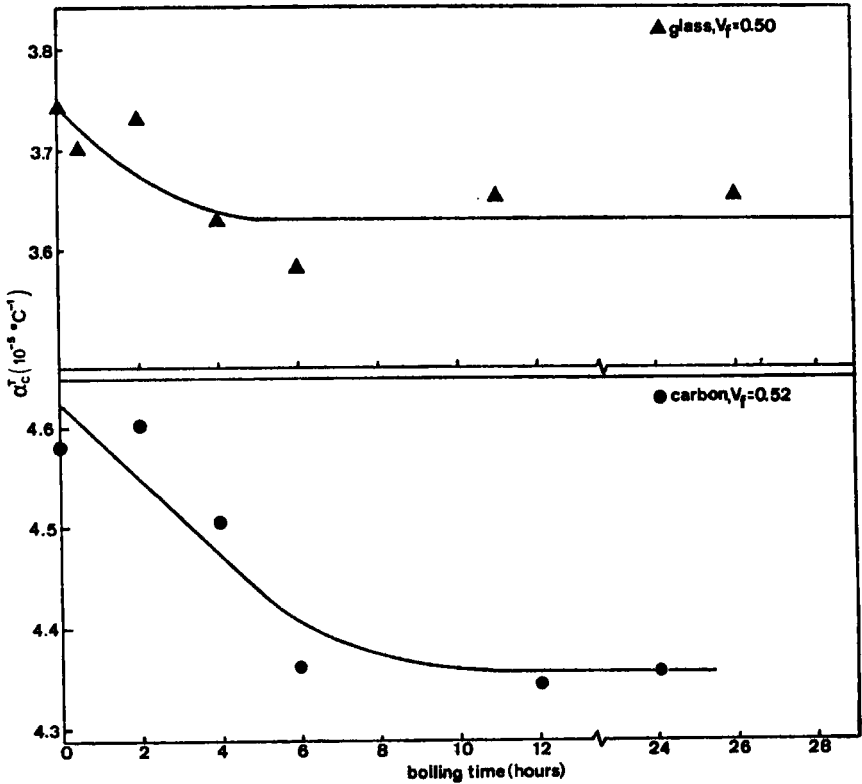


FIGURE 3 Results of transverse coefficients of thermal expansion as a function of boiling period.

Figure 2 indicates that a higher glass fibre volume fraction results in a decrease in the values of the longitudinal expansivity. A comparison between the glass fibre and the carbon fibre data shows that at close values of fibre volume fractions the first type of composite exhibits higher longitudinal expansivities and lower transverse expansivities compared with the latter type.

4. DISCUSSION

The trend of the expansivities as a function of the water-boil period is analysed first. A recent study by Marom and Weinberg⁴ pointed out the similarity between thermal expansion and other mechanical properties in the sense that they depended on the fibre matrix shear transfer mechanism. It was shown that thermal expansivities depended on the length of the fibre even in continuous fibre composites. A reduction in the fibre length resulted in an increase in the longitudinal expansivity and a decrease in the transverse one. The results were explained by applying the familiar efficiency factor, k (see for example Ref. 5) describing the effectiveness of the reinforcement, to the phenomenon of thermal expansion. It was proposed that the original equations derived by Schapery⁶ may be modified to express longitudinal and transverse expansivities (denoted by $\alpha_c'^L$ and $\alpha_c'^T$, respectively), which depend on the interfacial shear transfer as follows:

$$\alpha_c'^L = \frac{E_m \alpha_m V_m + k E_f \alpha_f V_f}{E_m V_m + k E_f V_f} \tag{1}$$

$$\alpha_c'^T = (1 + \nu_m) \alpha_m V_m + (1 + \nu_f) \alpha_f V_f - \alpha_c'^L (\nu_f V_f + \nu_m V_m) \tag{2}$$

Where E is Young's modulus, α is the coefficient of thermal expansion, V is the volume fraction, ν is Poisson ratio, and the subscripts m and f denote matrix and fibre properties, respectively. Equations 1 and 2 provided a reasonable explanation to the dependence of the experimental expansivities on the fibre length.⁴

The value of k is determined by the ratio of the fibre length to the fibre critical length, and it is given by the relationships:

$$k = l/2l_c \quad (l \leq l_c) \tag{3}$$

$$k = 1 - l_c/2l \quad (l \geq l_c) \tag{4}$$

Considering that l_c is given by the expression $l_c = r\sigma_f/\tau_i$, where r is the fibre radius, σ_f is its ultimate strength, and τ_i is the interfacial shear strength, it is seen how Eqs. 1 and 2 account for the effect of the fibre-matrix bond strength on the thermal expansivities. From Eq. 1 it is seen that a reduction in τ_i will result in an increase in $\alpha_c'^L$, while Eq. 2 shows that such a reduction will cause a decrease in $\alpha_c'^T$.

The possibility of utilizing thermal expansion measurements for determining τ_i depends on the l_c value produced by these measurements. It was shown⁴ that the value of l_c for the glass-epoxy system as calculated from thermal expansivity measurements was ~ 20 mm. It was also pointed out that this value corresponded with that of 13 mm found by measurements of strength of discontinuous glass fibre-reinforced epoxies.⁷ However, the values of τ_i

calculated from these l_c values are some two orders of magnitude smaller than those of 20–40 MN/m², usually measured in shear tests (see for example Ref. 8). It was reported that this discrepancy results from the fibres bunching together to give a much higher effective fibre diameter, hence the value of r used in the calculation of τ_i should be replaced by a higher effective radius.⁷ In view of the difficulties in assessing this effective radius or the extent of fibre interactions it is maintained that thermal expansivity measurement cannot provide a direct method for assessing the strength of the fibre-matrix bond. However, it may be used in a qualitative study of the process of fibre-matrix bond weakening.

The present work is an example for such a qualitative study. It discovers that the major deterioration of the fibre-matrix bond occurs during the first 5 hours of the exposure to boiling water, while later the process is very slow. This is true for both the glass fibres, which are chemically bonded to the epoxy matrix, and for the carbon fibres, which only form physical and mechanical ties.

The original expansivities of the unboiled specimens are discussed next. It can be shown that there is a reasonable agreement between these values and those calculated from Eqs. (1) and (2) ($k = 1$). The calculated values for the glass fibre composites are $\alpha_c^L(V_f = 0.37) = 0.9 \times 10^{-5}$, $\alpha_c^L(V_f = 0.55) = 0.7 \times 10^{-5}$, and $\alpha_c^T(V_f = 0.50) = 4.2 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$. The calculated values for the carbon fibre composites are $\alpha_c^L(V_f = 0.52) = 0.1 \times 10^{-5}$, and $\alpha_c^T(V_f = 0.52) = 5.8 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$. The following parameters were taken in the calculation:

epoxy matrix	glass fibre	carbon fibre ⁹
$E_m = 3.0 \text{ GN/m}^2$	$E_f = 70 \text{ GN/m}^2$	$E_f = 258 \text{ GN/m}^2$
$\nu_m = 0.35$	$\nu_f = 0.2$	$\nu_f = 0.26$
$\alpha_m = 6.0 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$	$\alpha_f = 0.5 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$	$\alpha_f^L \simeq 0$
		$\alpha_f^T = 2.9 \times 10^{-5} \text{ } ^\circ\text{C}^{-1}$

Equation (1) (for $k = 1$) also indicates that a reduction in V_f results in an increase in the value of α_c^L as in the case of the glass fibre composites. The longitudinal expansivity of the carbon fibre composite is in agreement with values quoted in Ref. 10 for the same temperature range, but slightly higher than the negative value quoted in Ref. 11. The transverse expansivity of this system is in the range given in Ref. 11 but higher than values given in Ref. 10. The deviations may be explained by different V_f values and different epoxy resins.

The difference between the results of the glass fibre composites and the carbon fibre composites is derived from the anisotropic nature of the carbon fibre. This anisotropy confers a very low coefficient of expansion in the fibre direction, but a relative high coefficient in the perpendicular direction.

Thus, the longitudinal expansivities of the carbon fibre composites are lower compared with those of the glass fibre materials, and vice versa for the transverse values.

5. SUMMARY OF CONCLUSIONS

1) Fibre-matrix bond strength affects thermal expansivities of composites. The effect of the weakening of this bond is to increase the longitudinal expansivities and to decrease the transverse expansivities.

2) The proposed theoretical expressions may explain these effects, though τ_i values calculated by these expressions are low compared with values measured directly.

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