JOURNAL OF MATERIALS SCIENCE 24 (1989) 1835–1844

Stress transfer in single-fibre composites: effect of adhesion, elastic modulus of fibre and matrix, and polymer chain mobility

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The stress transfer in single-fibre composites is studied experimentally by determining the critical fibre length to diameter ratio, l_c/d, in carbon fibre–epoxy resin or poly (ethylene vinyl acetate) systems. Our results and a great number of others available in the literature are compared with the predictions given, on the one hand, by the analytical approach by Cox and, on the other hand, by the theoretical study using finite element technique by Termonia. First, the influence of the fibre–matrix adhesion is analysed and it is observed, in agreement with Termonia, that l_c/d strongly decreases when the bonding efficiency between the two components is increased. Secondly, assuming a perfect fibre–matrix adhesion, it is shown that the critical fibre aspect ratio is proportional to the square root of the ratio of fibre to matrix elastic modulus, as predicted by Cox. However, two linear relationships are established: the first corresponds to the thermosetting and thermoplastic matrices, while the second corresponds to the elastomeric matrices. The difference between these two kinds of materials is attributed to the great difference in polymer chain mobility as shown by a study of the temperature dependence of l_c/d, particularly in the glass transition temperature zone of the matrices. However, in the case of elastomeric materials, the existence of an interphase layer between the fibre and the matrix, having an elastic modulus close to that of the elastomer in its glassy state, can also explain this particular behaviour.

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1. Introduction

There has been a rapid growth in the use of fibre-reinforced materials in engineering applications in the last few years, owing to their great versatility and high performance. These materials often consist of discontinuous stiff fibres embedded in a soft matrix with the fibre axis generally oriented in the direction of the applied load. In any fibre–reinforced resin, the load working on the composite is transmitted to the fibre through the fibre–matrix interface. Consequently, the structure and the properties of the fibre–matrix interface play a major role in the mechanical and physical properties of composite materials. In particular, the fibre–matrix interfacial shear strength is one of the most important parameters in controlling the toughness and the strength of a composite material. Its value is particularly dependent on any modification (fibre surface treatment, sizing, etc.) affecting the properties of the fibre–matrix interface.

The problem of stress transfer from the matrix to the fibre, across the interface, has recently received considerable attention. The model system which has been mainly considered is that of a single fibre embedded in a matrix under stress. The fragmentation test on single-fibre composite is suitable to measure directly the interfacial shear strength. A tensile load, applied to the specimen, is transmitted from the matrix to the fibre and, as originally described by Kelly and Tyson [1], the fibre should break into fragments until a limiting fragment size, defining a critical length l_c, is reached.

From a theoretical point of view, this system can be analysed by the classical shear lag theory, proposed by Cox [2], which permits analytical determination of the tensile stresses in the fibre and the shear stresses at the fibre–matrix interface in the case where both the fibre and the matrix are in the elastic state. The simple equations which resulted from this model are generally very useful for numerous applications. Unfortunately, Cox's analysis neglects the adhesion across the end faces of the fibres and fails to take into account local stress concentration effects near fibre ends. The importance of these assumptions has been demonstrated by finite element approaches [3]. In a recent paper, Termonia [4] has analysed theoretically the transfer of stress between the matrix and fibre in

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single-fibre composite, using finite difference technique. In particular, he showed that, at constant fibre diameter, the critical length is linearly related to the ratio of fibre to matrix modulus, in contradiction with Cox's model which predicts that, to a first approximation, l_c varies as the square root of this ratio. Nevertheless, all these predictions, from Cox's model as well as from Termonia's analysis, are based on the assumption that a perfect bond exists between the fibre and the matrix.

The aim of the present paper is first to determine qualitatively the influence of fibre–matrix adhesion on the critical length and to compare this influence with the theoretical finite element analysis presented by Termonia [4]. Secondly, considering a perfect bond between the fibre and the matrix, from our results and numerous others available in the literature, a general trend of variation of l_c in relation with the ratio of fibre to matrix modulus is examined, in order to determine which theoretical analysis, Cox's or Termonia's, is the most accurate one to describe the stress transfer from the matrix to the fibre in a single-fibre composite, whatever the nature of the fibre and the matrix.

The properties of resins which form the matrix are sensitive to temperature and it is conceivable that the critical length will be affected by temperature. Accordingly, in the last part of this study, the temperature dependence of the critical length is studied for carbon fibre–epoxy and carbon fibre–poly (ethylene vinyl acetate) composites, more particularly in the glass-transition temperature zone of the matrix.

2. Theory

In 1952, Cox [2] presented a theory applying to the case where both the fibre and the matrix are in the elastic state. The following major assumptions were made: (i) the fibre is surrounded by a cylindrical matrix; (ii) the transfer of load from matrix to fibre depends upon the difference between the actual displacement at a point on the interface, at a distance x from the end of the fibre, and the displacement that would be observed if the fibre were absent; (iii) a perfect bond exists between the fibre and the matrix; (iv) there is no load transfer through the ends of the fibre.

Considering a fibre length l embedded in a matrix under a general strain, ε, the following expressions for tensile stress, σ_f, in the fibre and shear stress, τ, along the fibre–matrix interface are then obtained

$$\sigma_f(x) = \varepsilon_f E_f = (E_f - E_m)\varepsilon\left[1 - \frac{\cosh \beta(l/2 - x)}{\cosh \beta\, l/2}\right] \quad (1)$$

$$\tau(x) = \frac{d\beta\sigma_c}{4}\left(\frac{E_f - E_m}{E_m}\right)\frac{\sinh \beta(l/2 - x)}{\cosh \beta l/2} \quad (2)$$

where E_f and E_m are the elastic moduli of the fibre and the matrix, respectively, ε_f is the axial fibre strain, σ_c the uniform stress applied to the composite, d the fibre diameter and β a constant defined as

$$\beta = \frac{2}{d}\left[\frac{E_m}{(1 + \nu_m)(E_f - E_m)\ln(2r_m/d)}\right]^{1/2} \quad (3)$$

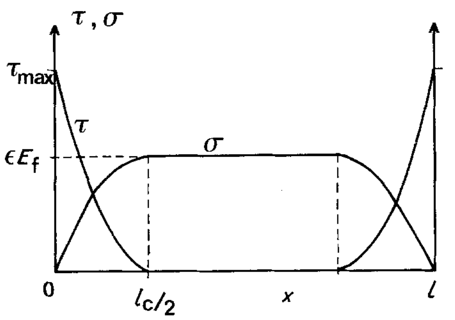

Figure 1 Theoretical variations of tensile (σ) and shear (τ) stresses along a fibre for $l > l_c$.

with ν_m the Poisson's coefficient of the matrix and r_m the radius of cylindrical matrix (or the separation between parallel fibres in a composite).

The expressions for σ_f and τ (Equations 1 and 2) give the usual longitudinal stress pattern (Fig. 1) which shows maximum fibre axial stress in the region of the midpoint of the fibre and maximum shear at the fibre ends [5]. This maximum shear strength, denoted τ_{max} (Fig. 1), for a fibre with a length equal to its critical length, can be written

$$\tau_{max} = \frac{d\beta}{4}\left(\frac{\sinh \beta l_c/2}{\cosh \beta l_c/2 - 1}\right)\sigma_f(l_c) \quad (4)$$

where $\sigma_f(l_c)$ is the tensile strength of this fibre at a length $l = l_c$.

It must be noted that the critical length is approximately equal to twice the distance between a fibre end (where $\sigma_f = 0$) and the point where σ_f reaches the maximum possible value of stress in fibre, i.e. εE_f, provided that the length l of the fibre is sufficient to obtain a variation of stress as given by Equation 1 having a maximum value of εE_f.

As stated by Galiotis *et al.* [6], for large values of l, Equation 1 can be simplified leading to the following form

$$\varepsilon_f/\varepsilon_m \simeq 1 - \exp(-\beta x) - \exp[-\beta(l - x)] \quad (5)$$

if $E_f \gg E_m$. It can be observed that for positions well away from the fibre ends $\varepsilon_f = \varepsilon_m$, while ε_f decreases exponentially to zero as x tends towards zero. The ratio $\varepsilon_f/\varepsilon_m$ reaches the value of $(1 - 1/e)$ at $x = 1/\beta$ or $x = 1 - l/\beta$. Thus, Galiotis *et al.* [6] consider that the inverse of the constant β can be identified, to a first approximation, as being equivalent to half the critical length previously defined. Consequently, from Equation 3 it is possible to write

$$\frac{l_c}{d} \simeq \left[\frac{(1 + \nu_m)(E_f - E_m)}{E_m}\ln\left(\frac{2r_m}{d}\right)\right]^{1/2} \quad (6)$$

and for $E_f \gg E_m$

$$\frac{l_c}{d} \simeq \left(\frac{E_f}{E_m}\right)^{1/2}\left[(1 + \nu_m)\ln\left(\frac{2r_m}{d}\right)\right]^{1/2} \quad (7)$$

In Equation 7, the term $[\ln(2r_m/d)]^{1/2}$ does lead to a logarithmic divergence when $r_m \to \infty$. Nevertheless, in a single-fibre composite, r_m is often taken equal to half the composite thickness considering that this value is in agreement with Cox's analysis. Fortunately, this term is a very slowly varying function, even for large values of the ratio r_m/d and, then, for a great number of fibre–matrix systems (considering also that the extreme values of $(1 + \nu_m)^{1/2}$ are equal to 1 and 1.225), it can be assumed that, in practice

$$\frac{l_c}{d} \simeq k \left(\frac{E_f}{E_m}\right)^{1/2} \tag{8}$$

where k is approximately constant. The scatter on the value of k will be discussed in Section 3.

Equation 8 must be compared with the results obtained by Termonia [4] using a finite element analysis. In this work, it was found that

$$\frac{l_c}{d} \simeq k' \frac{E_f}{E_m} \tag{9}$$

in contradiction with Equation 8.

It is also worth presenting another theoretical analysis given by Kelly and co-workers [1, 7] for the case of the matrix being in the plastic state. These authors have related the critical length, l_c, to the interfacial shear strength through a force balance equation around the embedded fibre. They assumed that the shear strength at the interface, τ, is constant and equal to the shear yield strength of the matrix and obtained the following equation

$$\tau = \frac{d}{2l_c}\sigma_f \tag{10}$$

where σ_f is the tensile strength of the fibre. This approach was analysed again by Di Benedetto and co-workers [8, 9] in a more general case. They found that

$$\tau = \frac{d}{2l_c}\sigma_f(l_c) \tag{11}$$

which is similar to the equation of Kelly *et al.*, but considers that $\sigma_f(l_c)$ is the tensile strength of the fibre at a gauge length equal to the critical length, l_c.

Recently, Simon and co-workers [10, 11] have shown that τ obtained by Equation 11 can be considered as the mean shear strength at the interface, because it is equal to the mean value of the shear stress $\tau(x)$ previously determined by Cox (Equation 2) according to

$$\tau = \frac{2}{l}\int_0^{l/2} \tau(x)\,dx \tag{12}$$

3. Experimental procedure

Two types of matrix were employed.

(1) A thermosetting epoxy resin: DGEBA–DDS (Ciba-Geigy LY 556, hardener HT 976). After adding the hardener (26% wt/wt) to the resin at 130° C, the mixture was stirred thoroughly for 15 min, defoamed in vacuum for 10 min, then poured into a mould, cured at 130° C for 3 h and post-cured at 180° C for 3 h under a pressure of 1.5 MPa. Sheets of about 2 mm thickness were then obtained.

(2) A thermoplastic resin: poly(ethylene-vinyl acetate) (EVA) (Elvax 150. Du Pont – 33% vinyl acetate content) was compression moulded (1.5 MPa), to obtain 2 mm thick sheets in a picture frame type mould at 160° C for 2 min and then quenched to room temperature.

Tensile specimens according to ISO 1/2 standard were cut from the sheets in both cases. To determine the mechanical behaviour of both matrices, in particular their elastic modulus E_m, stress–strain curves were obtained on an Instron 1195 H tensile testing machine at a constant cross-head speed equal to 0.5 mm min^{-1}. The testing machine was equipped with an environmental chamber enabling temperatures to be varied for the epoxy resin from room temperature to 180° C and for EVA from -60 to $+40$° C.

The glass transition temperature, T_g, of the matrices was determined by differential scanning calorimetry (DSC) with a Mettler TA 3000 apparatus. The DSC spectra were recorded with an increasing temperature rate of 10° C min^{-1}. The values of T_g were found equal to 140 and -36° C for epoxy resin and EVA polymer, respectively. It was also observed that the degree of crystallinity of EVA was low, less than about 10%.

The fraction of insoluble material (gel) present in the EVA polymer was determined by immersing about 1 g EVA in toluene for 7 days at room temperature. After filtration, washing with toluene and drying overnight at 40° C under vacuum, the insoluble fraction of the polymer was found equal to about 9%. This very low value shows that there is practically no permanent network and therefore the degree of cross-linking of EVA is small.

The fibres used in this study were high-strength PAN-based carbon fibres having received a surface treatment through electrolytic oxidation [12] in order to strongly increase the adhesion at the fibre–matrix interface. Their mechanical properties were determined on monofilaments at different gauge lengths varying from about 5 to 70 mm on the Instron tensile machine at a cross-head speed of 0.5 mm min^{-1}. Their elastic modulus was found equal to 240 ± 15 GPa. Their tensile strength was dependent on the gauge length, l, and a linear relationship between $\ln(\sigma_f)$ and $\ln(l)$ was established

$$\ln(\sigma_f) = -0.17\ln(l) + 21.26 \tag{13}$$

This logarithmic dependence of σ_f with l allows us to extrapolate the value of $\sigma_f(l)$ at the critical length l_c (and then to calculate both τ_{max} and τ by Equations 4 and 11). l_c is usually too small, ranging from about 0.3 to 3 mm, to perform experimental measurements of individual fibre strengths.

For the fragmentation test, single-fibre composites were prepared by pouring the epoxy resin into a special mould holding ten carbon fibres in the centre of the sheet and then curing the composite as above. For EVA matrix, two sheets (1 mm thick) were previously obtained as above and moulded again with the same experimental conditions on each side of a picture frame also supporting ten carbon fibres. Specimens according to ISO 1/2 standard were cut carefully so that the fibre axis coincided with the axis of the composite as well as possible. Each sample was subjected, at a cross-head speed of 0.5 mm min^{-1} in the environmental

TABLE I Single-fibre composites for which results are taken into account in the present study

Matrix	Fibre	Reference
Unsaturated polyester	Glass	[13]
Epoxy resin	Glass	[13]
Epoxy resin	Substituted polydiacetylene	[6, 14]
Epoxy resin	Carbon	[12, 15, 16]
Epoxy resin	Graphite	[17, 18]
Polyethylene	Glass	[10, 11, 19]
Polyvinylchloride	Glass	[10, 11]
Ether block amide copolymers	Glass	[20]
Polypropylene	Glass	[21]
Polyurethane	Glass, carbon	[11, 22]

chamber of the testing machine, to a tensile strain greater than the fibre ultimate tensile strain, which was found equal to about 1.5%. After testing, the fibre in the composite was broken into a large number of fragments (20 to 50). Mean fibre fragment length, $\bar{l}$, was determined using an optical microscope equipped with a micrometer eyepiece. According to Kelly and Tyson [1], the critical length, l_c, is equal to $4/3\ \bar{l}$ because the length of broken fibre pieces should be distributed in the range $l_c/2$ to l_c.

A great number of results presented in this study come from a survey of the literature. Thus, the systems, for which accurate data are available, are given in Table I. It is shown that matrices are thermosetting or thermoplastic polymers as well as elastomers and that fibres are glass, carbon or organic fibres. For all these different cases, as well as for our own experiments, it appeared that the values of the term $[(1 + v_m) \ln (2r_m/d)]^{1/2}$ in Equation 7 were always included between 2 and 3, corresponding to $40 \lesssim 2r_m/d \lesssim 300$ and $0.3 \lesssim v_m \lesssim 0.5$. Therefore, the assumption concerning the coefficient k (Equation 8) and discussed in Section 2, is confirmed, k can be considered a constant with a mean value equal to 2.5.

4. Results and discussion

4.1. Effect of fibre–matrix adhesion on the critical length

The effect of the adhesion on the critical length has been studied theoretically by Termonia [4] using finite element analysis. He assumed that this adhesion can be described by an "adhesion factor" equal to one when all the possible bonds at the fibre–matrix interface are effective. The adhesion factor decreases when bonds are broken to reach the zero value at zero level of adhesion. Termonia observed that a decrease of the fibre–matrix adhesion leads to an increase of the critical length, this increase being particularly rapid when the adhesion factor becomes less than about 0.3 (see Fig. 6 in [4]).

Two experimental studies concerning the effect of adhesion on the fibre–matrix interfacial shear strength using a fragmentation test have been performed recently. The first one by Simon and co-workers [10, 11] concerns the adhesion between glass fibres and polyethylene. The glass fibres have been primarily submitted to different surface treatments (chemical etching treatment, silanes treatment, etc.). In Fig. 2 the variation of the critical aspect ratio, l_c/d, is plotted against the work of adhesion, W_a, determined by wettability techniques. In that case, because polyethylene is a non-polar material, only dispersive interactions are established between the fibre and the matrix. The curve obtained (Fig. 2) is qualitatively the same as that given by Termonia [4].

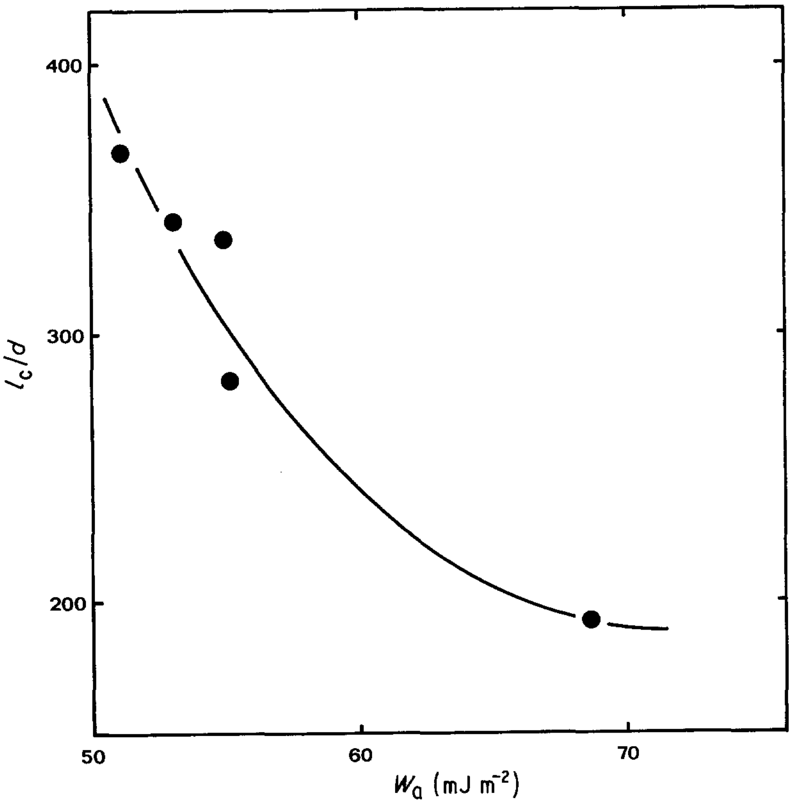

Figure 2 l_c/d plotted against work of adhesion, W_a, for a glass fibre–polyethylene system [10, 11].

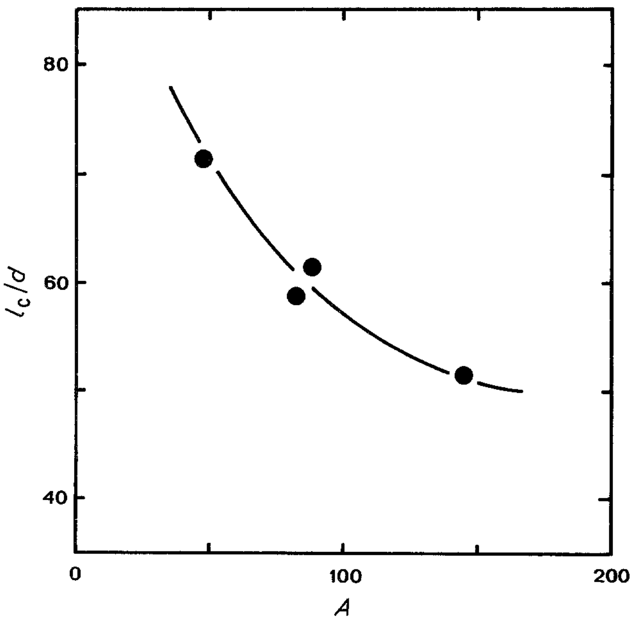

Figure 3 l_c/d plotted against specific interaction parameter, A, for a carbon fibre–epoxy resin system [12].

The second study concerns carbon fibre–epoxy resin systems [12]. As before, the characteristics of the matrix remained unchanged and the carbon fibres were surface treated by oxidation or sizing. In contrast with the previous study, both fibre and matrix are now polar materials and thus a specific interaction parameter, A, between these two constituents can be measured by means of inverse gas/solid chromatography technique [23]. This specific interaction parameter describes acid–base interactions [24] at the fibre–matrix interface. It appears also that the plot of l_c/d against A (Fig. 3) has the same qualitative form as that obtained in the previous study (Fig. 2) and in Termonia's work.

These results point out the great influence of the fibre–matrix adhesion on the value of the critical length which can be divided by a factor equal to about two when an appropriate surface treatment is applied to the fibre.

It is worth remembering now that in the theories of Cox [2] and Termonia [4] it is assumed that a perfect adhesion must exist between the fibre and the matrix to determine the magnitude of the stress transfer. Thus, for the continuation of this paper and particularly in the case of literature data concerning a given system, only the smallest available values of l_c corresponding to the highest values of interfacial shear strength are taken into account. Moreover, it will be assumed that in each case the best level of fibre–matrix adhesion is reached. It is also the reason why oxidized carbon fibres are used in our own experiments, considering that the oxidation treatment is efficient enough to create a very good adhesion between the fibres and the epoxy resin or the EVA polymer.

4.2. Relationship between l_c/d and E_f/E_m

In order to determine which theoretical analysis, that of Cox (Equation 8) or that one of Termonia (Equation 9), is the most accurate to describe the stress transfer from the matrix to the fibre in a single-fibre composite, the variation of l_c/d with the ratio E_f/E_m, fibre to matrix modulus in logarithmic scales, is shown in

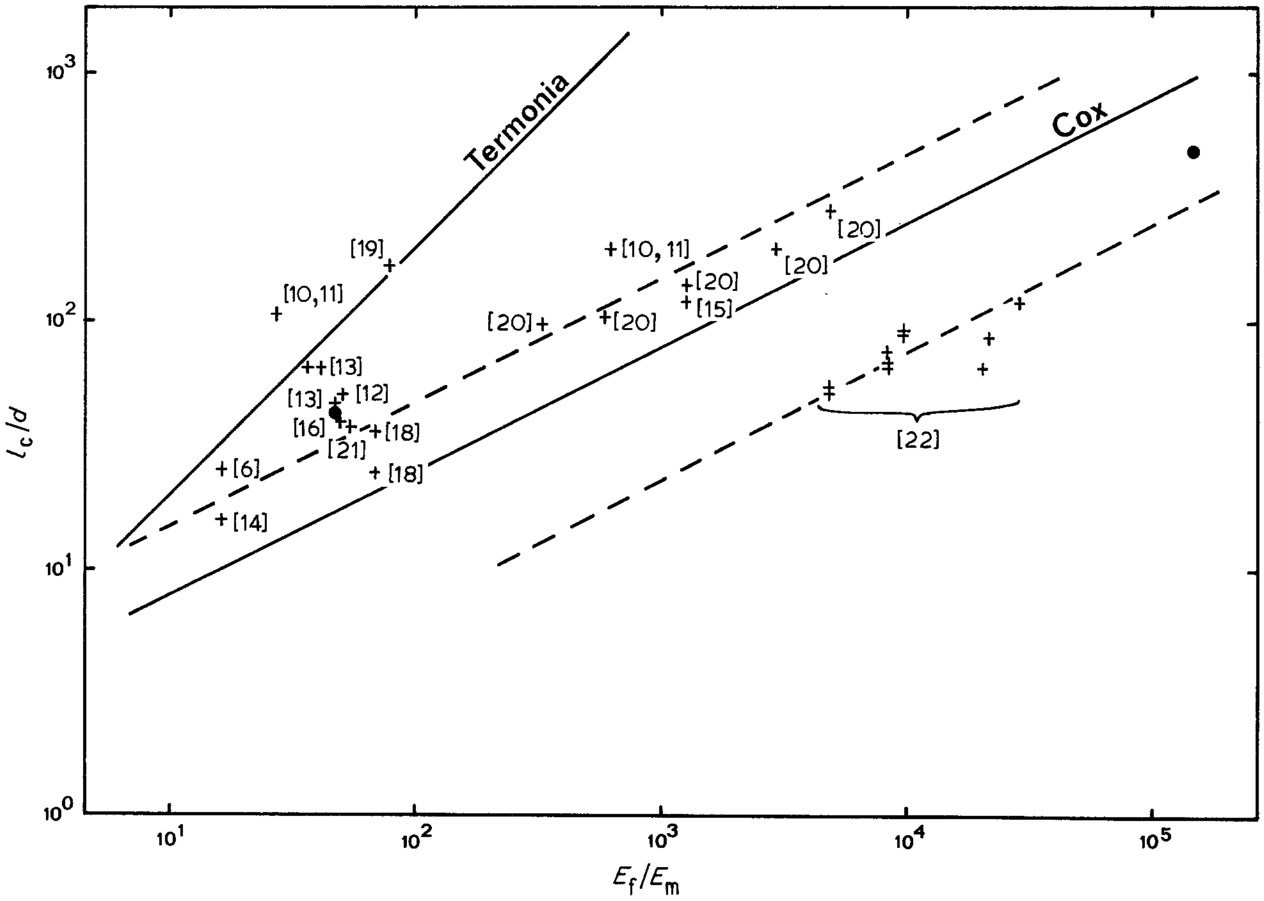

Figure 4 l_c/d plotted against E_f/E_m in logarithmic scales for different systems: (●) present results, (×) results from the literature (numbers near the points correspond to the references); (——) theoretical predictions, (– – –) experimental fittings.

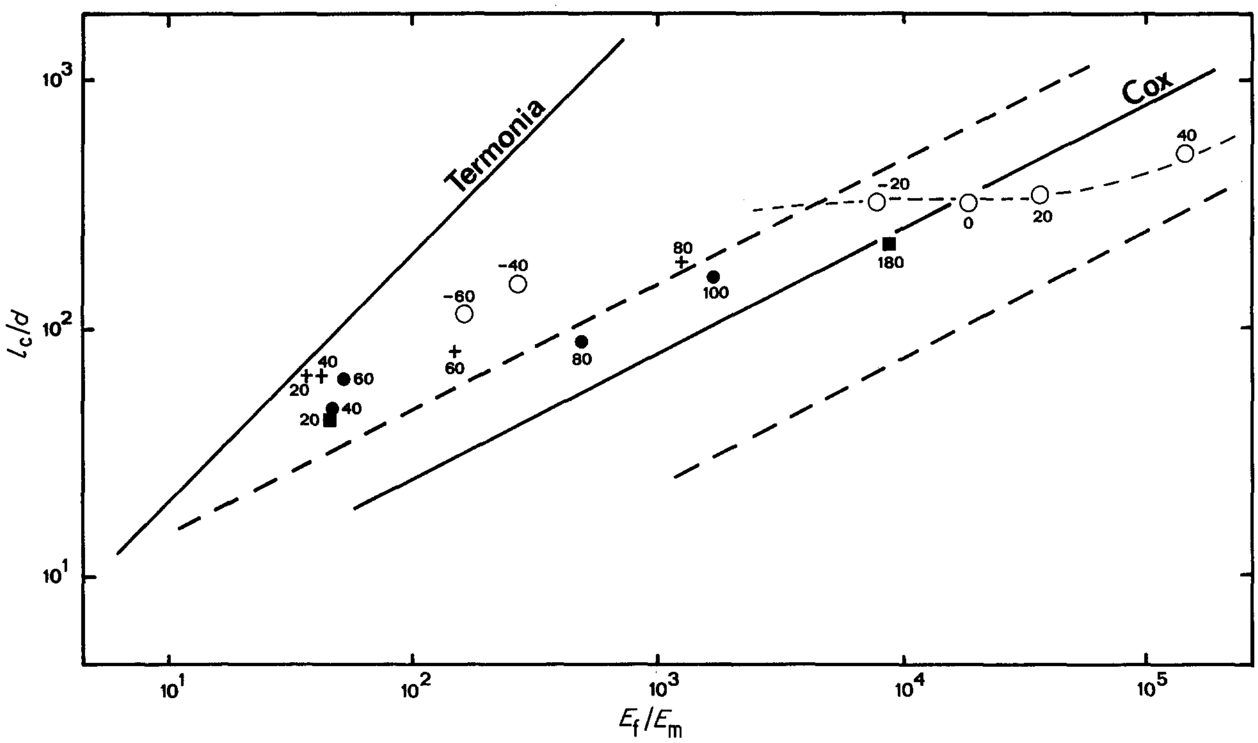

Figure 5 Influence of the temperature on the variation of l_c/d with E_f/E_m in logarithmic scales for different systems: (●) glass fibre–unsaturated polyester [13], (×) glass fibre–epoxy resin [13], (■) carbon fibre–epoxy resin, (○) carbon fibre–EVA, (——) theoretical predictions, (– – –) experimental fittings (see Fig. 4). (Numbers near the points correspond to the temperatures in °C).

Fig. 4, where the theoretical straight lines corresponding to Cox and Termonia's models are also drawn.

It immediately appears that only four or five experimental points agree with Termonia's analysis, especially for low values of E_f/E_m. On the contrary, all the other results (27 experimental values) are located on straight lines which are parallel to that of Cox. A first set of values, corresponding to thermosetting and thermoplastic matrices only, defines a linear relationship between log (l_c/d) and log (E_f/E_m) having a slope equal to about 0.5 in agreement with Equation 8 (the best fit gives a slope equal to 0.44 ± 0.04). Nevertheless, all these values are located above Cox's theoretical line and it is possible to write

$$l_c/d \simeq 4.7(E_f/E_m)^{1/2} \tag{14}$$

This equation has, of course, the same mathematical form as Equation 8. However, the value of the coefficient k, experimentally equal to 4.7 in this case, is higher than the predicted one, $k \simeq 2.5$, as defined in Section 3. One of the major assumptions made by Cox is that a perfect adhesion exists between the fibre and the matrix. Experimentally, it is very difficult to check this assumption. Moreover, as previously observed, the ratio l_c/d greatly increases when the fibre–matrix adhesion is decreased. Therefore, a small lack of adhesion can explain this experimental value of k. Such a phenomenon could also be put forward for the few points which are in agreement with Termonia's relationship.

A second set of experimental values, corresponding to matrices exhibiting a viscoelastic behaviour (polyurethane and EVA), again defines a linear relationship also having a slope of about 0.5 (best fit 0.58 ± 0.08), but this straight line is now located below that of Cox, because the coefficient k is found equal to about 0.65, according to

$$l_c/d \simeq 0.65(E_f/E_m)^{1/2} \tag{15}$$

Remembering that in order to correlate l_c/d with E_f/E_m it has been assumed that $1/\beta \simeq l_c/2$, Equations 14 and 15 lead, in fact, to the following equalities:

for thermosetting–thermoplastic materials $1/\beta \simeq 0.5\,l_c/2$

for elastomeric materials $1/\beta \simeq 3.8\,l_c/2$.

Finally, whatever the nature of the fibres, it seems that the stress transfer from an elastomeric matrix to these fibres is far better than from a thermosetting or a thermoplastic matrix in all cases. For a given value of the ratio E_f/E_m, the mean value of l_c/d is about seven times smaller for an elastomeric material than for another matrix. It is concluded that the nature of the matrix plays an unexpected major role in the process of stress transfer through the interface. Nevertheless, for the greater part of results, as stated by Cox, the stress concentration near fibre ends in a single-fibre composite can be neglected.

Three fundamental phenomena can be invoked in order to explain the results in the case of elastomeric materials:

(i) a frictional effect due to the transverse contraction of the incompressible elastomeric matrix ($\nu_m = 0.5$). At high applied strain, this effect can be responsible for an important additional stress acting on the fibre and then reducing its critical length;

(ii) a viscoelastic effect similar to that generally observed in several adhesion measurements and studied previously [25, 26]. In that case, it is probable that the stress distribution along the fibre is more homogeneous than for a pure elastic matrix;

(iii) in the case of elastomeric matrices, the existence of an interphase layer between the fibre surface and

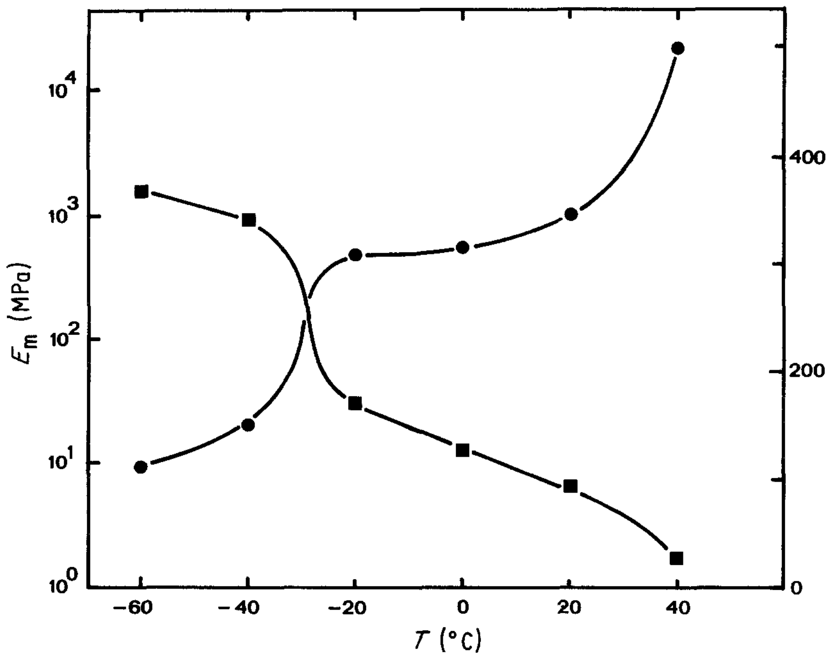

Figure 6 (■) Log (E_m) and (●) l_c/d plotted against temperature for the carbon fibre–EVA system.

the matrix. In this zone around the fibre, it can be considered that either the molecular packing density of the elastomer or the motion of polymer chains are quite different than those of the bulk matrix material. Therefore, it is conceivable that the elastic modulus of this layer is higher than that of the bulk matrix. Of course, for the fragmentation test, the elastic modulus of this interphase layer must be taken into account instead of that of the bulk material.

A few experiments have been performed in order to check the effect of the frictional force. The critical length of a carbon fibre embedded in EVA elastomer was determined at different levels of external strain at 20° C. It appeared that when the applied strain became equal to about three to four times the tensile ultimate strain of the fibre, the critical length of this fibre reached a minimum value which remained unaffected by extra strain. Therefore, it can be concluded that the frictional effect, although it actually exists, is totally uneffective to reduce the critical length of a fibre in elastomeric matrices, even at high values of external applied strain (50 to 100%).

This result allows us to emphasize the significance of the other phenomena listed above, i.e., the viscoelastic behaviour of the matrix and the existence of an interphase layer. Such effects are highly dependent on the temperature at which experimental measurements are performed. Consequently, in the next section, the temperature dependence of the critical length for our fibre–matrix systems is discussed, particularly near the glass transition temperature of the matrix.

4.3. Temperature dependence of l_c/d

To our knowledge, only one study (Ohsawa *et al.* [13]) has been devoted to the temperature dependence of critical fibre length measured by the fragmentation test. The authors have determined the variation of both elastic modulus of the matrix, E_m, and critical fibre length, l_c, as a function of the temperature, T, varying from 20 to about 120° C, for glass fibre-reinforced epoxy resin or unsaturated polyester. The glass transition temperature, T_g, was found to be about 60 and 40° C for epoxy resin and polyester, respectively. They found that the ratio l_c/d increases greatly but continuously with temperature, while the curves E_m against T dropped near to the T_g value of each matrix. Nevertheless, by plotting log (l_c/d) against log (E_f/E_m) (Fig. 5) for both systems studied by Ohsawa *et al.* [13] it appears that, whatever the temperature, below or above T_g, the experimental results agree rather well with the linear relationship obtained previously (Equation 12) and corresponding to the thermosetting and thermoplastic matrices. For low values of T, a few points are located near Termonia's relationship.

Therefore, for those kinds of systems, it seems that even at temperatures much higher than T_g it is difficult or impossible to obtain experimental values which agree with the straight line corresponding to elastomeric matrices. It is worth noting that materials used by Ohsawa *et al.* [13], namely epoxy and polyester resins, are highly cross-linked polymers. This fact allows us to think that certainly the mobility of polymer chains is a major factor influencing the critical fibre length in single-fibre composites.

In Fig. 5, it can also be observed that similar results were obtained for our carbon fibre–epoxy resin system (T_g = 140° C), at 20 and 180° C. Nevertheless, the second value is intermediate between the two straight lines defined above and agrees quite well with Cox's theory. It seems that at higher temperatures, it could be possible to reach the straight line of elastomers; however, beyond 180° C a thermal degradation of epoxy resin occurs and it is therefore impossible to perform reproducible experiments.

For the carbon fibre–EVA system (T_g = −36° C) studied in this paper, the results are quite different. Fig. 6 shows simultaneously the variations of both E_m and l_c/d as a function of temperature, T. Contrary to the results of Ohsawa *et al.* [13], both variations present an inflexion point near the EVA T_g value. However, it appears in Fig. 5 that for T lower than T_g, the values of log (l_c/d) against log (E_f/E_m) agree rather well with the first straight line (thermosetting and thermoplastic materials) and for T much higher than T_g, the

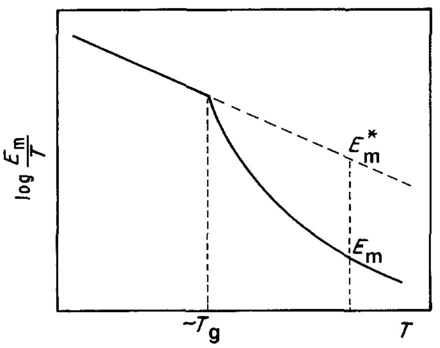

Figure 7 Schematic variation of log (E_m/T) with absolute temperature, T, and estimation of the elastic modulus E_m^*, of the interphase layer.

experimental results are located in the vicinity of the second straight line (elastomeric materials). In the intermediate region, it seems that the values of l_c/d are kept almost constant whatever the temperature.

By gel content and differential scanning calorimetry (DSC) measurements, we have noticed that EVA is a polymer with a low degree of cross-linking and crystallinity so that, at temperatures higher than its T_g, it can be considered as an elastomer, although it is often known as intermediate between a thermoplastic polymer and an elastomer [27, 28]. Therefore, at these temperatures, the mobility of the polymer chains is high, leading to a marked viscoelastic behaviour of the matrix. Finally, it clearly appears that the polymer chain mobility is a major factor determining the level of stress transfer at the fibre–matrix interface in fibre-reinforced composites.

If it is clear that for both cases (below and above T_g), variations of (l_c/d) against (E_f/E_m) in logarithmic scales are unique straight lines according to Equation 8, however, it is not easy to give an explanation for the values of the coefficient k in Equations 14 and 15. Finally, Equation 8 has to be corrected as follows

$$\frac{l_c}{d} = \alpha k_0 \left(\frac{E_f}{E_m}\right)^{1/2} \qquad (16)$$

where k_0 is equal to the theoretical mean value of the term $[(1 + v_m) \ln (2r_m/d)]^{1/2}$ determined in Section 3, i.e. $k_0 \simeq 2.5$, and α is a new coefficient which can take only two discrete values, α_{ther} and α_{elast}, corresponding, respectively, to thermosetting or thermoplastic polymers and elastomers. From Equations 14 and 15, we obtain the following experimental values: $\alpha_{\text{ther}} \simeq 2$ and $\alpha_{\text{elast}} \simeq 1/4$. These values of α can be related to the mobility of the polymer chains but it would be difficult to give a more precise physical meaning.

Let us assume now that in the case of elastomeric materials, an interphase layer exists between the fibre surface and the matrix. In this layer, a different molecular packing density of the elastomer than that of the bulk material can be considered and the motion of polymer chains is certainly restricted by the presence of the fibre. For highly cross-linked polymers and thermoplastics or elastomers in their glassy state, this phenomenon can be neglected. To a first approximation it can be assumed that the mechanical behaviour of this layer is equivalent to that of the elastomer in its glassy state, even at temperatures much higher than T_g.

In order to estimate the elastic modulus E_m^* of this interphase layer, we have checked for EVA elastomer that an absolute temperature dependence of E_m/T can be established, according to the work of Ilavsky and Hasa [29]. Therefore, as shown in Fig. 7, it is possible to determine E_m^* of the interphase layer by extrapolating at temperatures beyond T_g, the linear relationship observed between log (E_m/T) and T at temperatures lower than T_g. Calculated values of E_m^* are given in Table II.

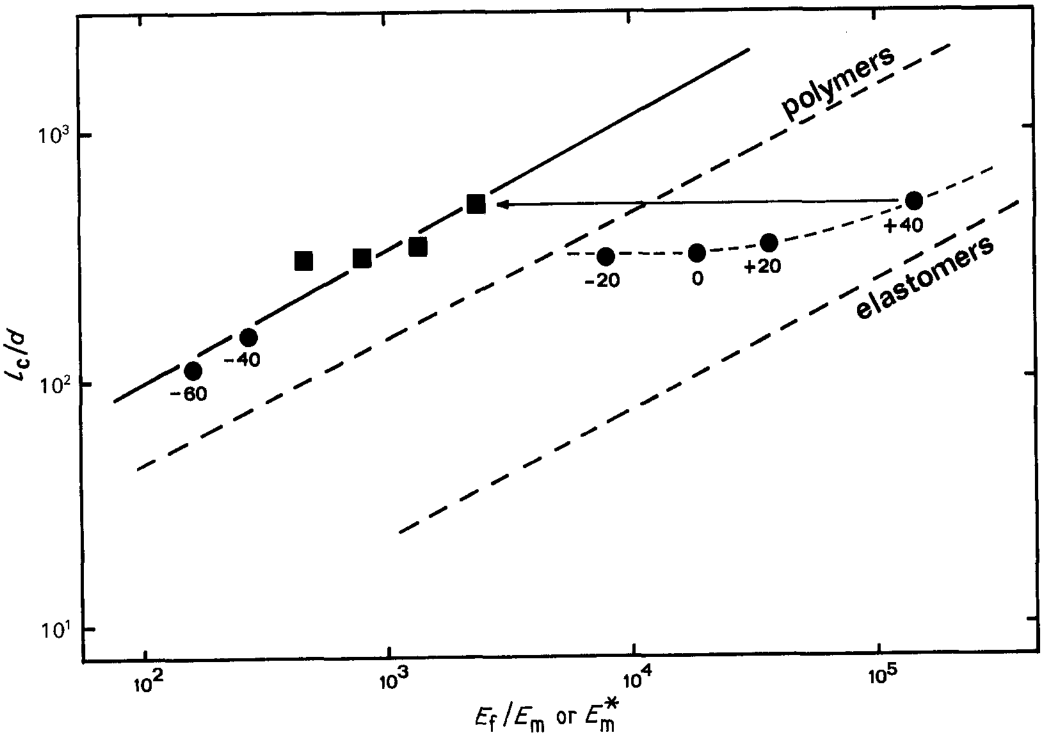

Figure 8 l_c/d plotted against (●) E_f/E_m or (■) E_f/E_m^* for EVA elastomer (numbers near the points correspond to the temperatures in °C).

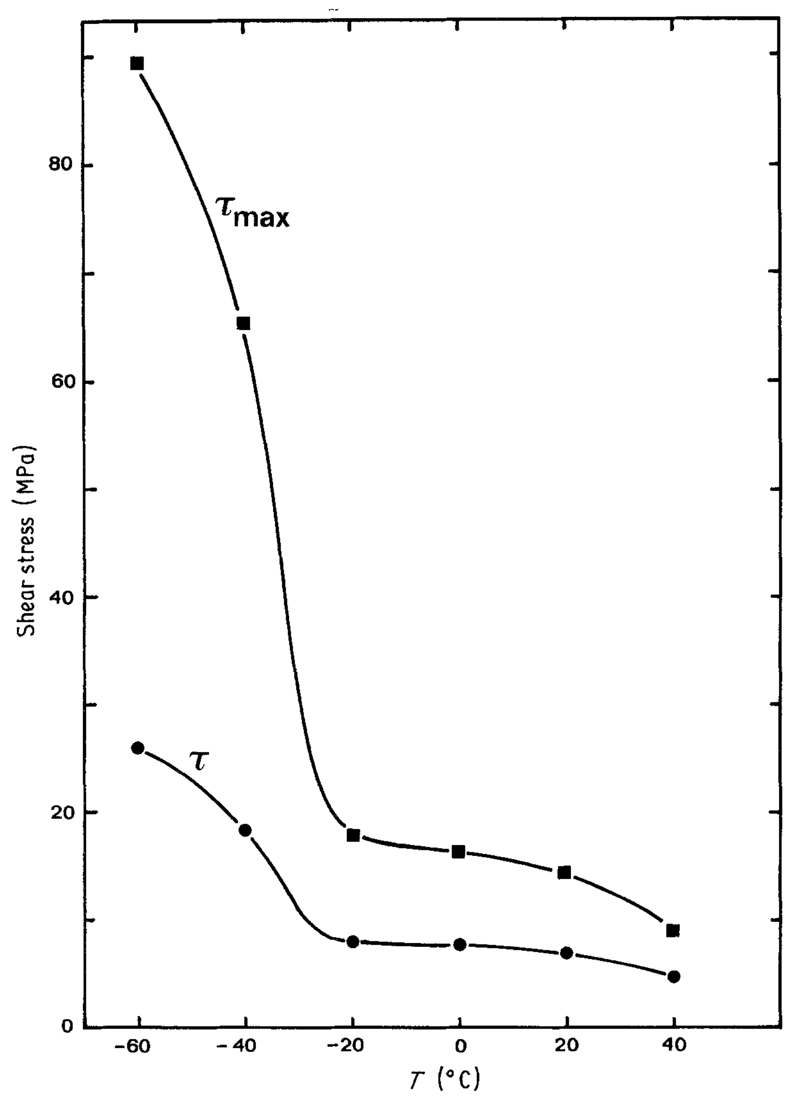

Figure 9 Interfacial shear stresses τ and τ_{max} plotted against temperature for carbon fibre–EVA system.

Taking into account the values of E_m^* instead of E_m for the EVA elastomer in the rubbery region, it immediately appears in Fig. 8 that all the values of l_c/d against E_f/E_m in logarithmic scales define a straight line, having a slope of about 0.5, quite close to the line corresponding to the thermoplastic and thermosetting matrices. The fact that the former is located above the latter can be explained by a lack of fibre–matrix adhesion as previously discussed.

Finally, the assumption of the existence of an interphase layer between the fibre surface and the elastomeric matrix could explain rather well the results of stress transfer in single fibre–elastomer composites.

For this carbon fibre–EVA system, it is now interesting to calculate according to Equations 4 and 11 the values of the maximum shear strength, τ_{max}, and the mean shear strength, τ, at the interface, respectively. The relationships between both shear strengths and the temperature are shown in Fig. 9. It can be observed that τ_{max} and τ decrease rapidly for temperatures lower than the T_g of the matrix, while beyond T equal to about $-20°$ C, both strengths decrease less steeply with increasing temperature. These types of variation are quite different from the linear decrease observed elsewhere [13] for systems involving cross-linked matrices.

The fact that shear strengths at the interface decrease with increasing temperature (as shown in Fig. 9) is certainly not brought about either by a decrease in bond strength at the fibre–matrix interface or by a thermal stress produced by the difference in thermal expansion coefficient between fibre and resin. It can, however, be explained by a decrease in shear strength of the matrix with increasing temperature [13]. However, observations under a microscope of the breaking point of a fibre in the samples submitted to the fragmentation test at $T > T_g$ do not reveal cracks in the matrix. Therefore, it can also be assumed that secondary bonds existing at the fibre–matrix interface undergo relaxation with increasing temperature and consequently, the shear strengths at the interface decrease. Nevertheless, the shape of the variations of τ_{max} and τ against T (Fig. 9) (this shape being greatly dependent on the glass transition temperature of the matrix), is evidence for the major influence of the shear strength of the matrix.

TABLE II Comparison between measured elastic modulus (E_m) of bulk elastomer and calculated elastic modulus (E_m^*) of the interphase layer, in the rubbery region of EVA matrix

T (°C)	E_m (MPa)	E_m^* (MPa)
−20	30.5	524
0	12.9	307
+20	6.6	179
+40	1.7	104

5. Conclusion

In this study it was shown, from our results and numerous others available in the literature, that the classical shear lag theory proposed by Cox [2], almost 40 years ago, can still interpret very well the stress transfer at the fibre–matrix interface in fibre-reinforced polymers. In particular, in single-fibre composite as stated by Cox, the stress concentration near fibre ends can be neglected in contradiction with recent theoretical analysis by means of finite element techniques [4].

First, the effect of fibre–matrix adhesion on the critical length was studied. It was observed as expected that the critical length increases with decreasing adhesion. Secondly, considering a perfect bond at the fibre–matrix interface, it was shown that the critical fibre aspect ratio (l_c/d) is linearly related to the square root of the ratio fibre to matrix modulus in agreement with Cox's theory, whatever the nature of both fibre and matrix. Nevertheless, two linear relationships were established: the first corresponds to the thermosetting and thermoplastic matrices and the second to the elastomeric matrices, both straight lines having slopes different from that predicted by the theory. Therefore, it is concluded that the mobility of the polymer chains can be taken as responsible for this experimentally observed behaviour. On the one hand, for highly cross-linked matrix (epoxy resin) it was observed that it is difficult to cross from the first relationship to the other one. On the other hand, for a matrix (EVA) having a low degree of cross-linking, it has been shown that the variation of the critical fibre aspect ratio for temperatures below the glass transition temperature of the matrix agrees with the first relationship, whereas this variation agrees with the second relationship at temperatures above T_g. Nevertheless, the assumption of the existence of an interphase layer between the fibre surface and the elastomeric matrix could explain these particular results of stress transfer in single fibre–elastomer composites. In this case, it is assumed that, whatever the temperature, the elastic modulus of the interphase layer is equal to that of the elastomer in its glassy state.

Acknowledgements

The authors thank Dr B. Haidar for helpful discussions and Dr G. Guilpain for personal communications of unpublished results.

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Received 28 March
and accepted 29 July 1988