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The strength of the fibre–polymer interface in short glass fibre-reinforced polypropylene

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A method for the determination of the interfacial bond strength in well-aligned short glass fibre-reinforced polypropylene samples is discussed. The method takes into account the variation of the interfacial shear stress during the deformation process; consequently, it yields very consistent results at all values of the composite strain. The influence of the fibre orientation with respect to the load axis is appropriately considered using macro-mechanical analyses for stiffness and strength of the composite. The method is compared with two other methods reported in the literature.

1. Introduction

The improvement in strength of short glass fibre-reinforced thermoplastics is greatly dependent upon the characteristics of the numerous fibre–matrix interfaces which exist in a composite sample. For a satisfactory transfer of stress between the polymer matrix and the fibre reinforcement, it is necessary that sufficient adhesive strength is developed along the fibre–polymer interface. A poorly bonded region at the interface will cause rupture of the interface at a very low shear stress. This debonded (dewetted) region will then spread along the interface until the whole fibre is separated from the matrix. Around the resulting cavity, high stress concentration can develop and this enhances the failure probability of the sample.

It is, therefore, useful to be able to determine quantitatively the interfacial bond strength, τ. Bowyer and Bader [1] and Ramsteiner and Theysohn [2] have suggested methods for this determination. Both these methods are based on the fibre pull-out model of Kelly and Tyson [3]. Bowyer and Bader's method allows the determination of τ from studies on a composite sample containing discontinuous fibres and is based on the assumption that τ is constant at all composite strains. The method suggested by Ramsteiner and Theysohn is based on a linear relationship between the strength of the composite and the fibre volume fraction. It requires the measurement of the strengths of samples having well-aligned discontinuous fibres with different volume fractions and is therefore necessarily time consuming.

In the present work, we use the stress–strain relation of a single sample of the composite with fibres aligned along the load axis in order to determine τ at failure. The strengths and moduli of samples having different orientation to the load axis are shown to be consistent with macro-mechanical theories.

2. Theory

The starting point is the fibre pull-out model of Kelley and Tyson [3]. According to this model, there is a critical length, l_c, given by the following expression:

$$l_c = \frac{E_f r_f \epsilon_c}{\tau} \tag{1}$$

where E_f is Young's modulus for the fibre material, r_f is the fibre radius, ϵ_c is the composite strain and τ is the interfacial shear stress. For a subcritical fibre of length l, the average stress is:

$$(\bar{\sigma}_f)_{sub} = \frac{l\tau}{2r_f}, \tag{2}$$

and for a supercritical fibre of length l, the average stress is:

$$(\bar{\sigma}_f)_{sup} = E_f \epsilon_c \left(1 - \frac{E_f \epsilon_c r_f}{2l\tau}\right). \tag{3}$$

Using the well-known law of mixtures for composites, the composite stress can be calculated. Bowyer and Bader [1] suggested the final expression as:

$$\sigma_c = \eta(X+Y)+Z, \tag{4}$$

where

$$X = \Sigma_{(\text{subcritical})}\, \tau L_i V_i/2r_f, \tag{5}$$

$$Y = \Sigma_{(\text{supercritical})}\, E_f \epsilon_c \left(1 - \frac{E_f \epsilon_c r_f}{2L_j \tau}\right) V_j, \tag{6}$$

and

$$Z = E_m \epsilon_c (1 - V_f), \tag{7}$$

where η is the orientation factor; V_i and V_j are the volume subfractions of fibres with lengths l_i and l_j, respectively, E_m is the matrix modulus and V_f the volume fraction of all fibres. In Equation 5, the summation extends over all fibre-length intervals below the critical length and in Equation 6 over all intervals above the critical length.

While applying the equations given above, Bowyer and Bader assumed that the value of the shear stress (τ) is same at all values of the composite strain (ϵ_c). The second assumption made by them was that the Kelly and Tyson model can be applied at all fibre orientations to the load axis by incorporating an orientation factor, η and thus an imperfectly oriented sample can be analysed by their procedure and a representative value of η obtained. It is shown below that the constancy of τ, particularly at low values of ϵ_c, leads to widely varying values of η for the same sample.

In contrast to Bowyer and Bader's procedure, Ramsteiner and Theysohn used well characterized and aligned composite samples with fibres oriented along the load axis. But they assumed that the volume fraction of supercritical fibres was negligible for their samples and hence $Y=0$ in the above equations. τ was obtained from the slope of curves between the composite stress at failure and the fibre volume fraction.

In order to correlate the stiffness and strength of aligned fibre samples with different orientations to the load axis, it is essential to use macromechanical analysis (see, for example, [4]). Accordingly, the Young's modulus, E_θ for an orthotropic material loaded in uniaxial tension or compression along an axis inclined at angle θ to the fibre direction is given by the following relation:

$$\frac{1}{E_\theta} = \frac{\cos^4\theta}{E_1} + \left(\frac{1}{G_{12}} - \frac{2\nu_{12}}{E_1}\right)\sin^2\theta\cos^2\theta + \frac{1}{E_2}\sin^4\theta, \tag{8}$$

where E_1 and E_2 are, respectively, the longitudinal and transverse moduli, G_{12} the shear modulus and ν_{12} the Poisson's ratio of the orthotropic sheet in the principal material co-ordinates.

Similarly the variation of tensile strength, σ_θ, with θ is examined using the Tsai–Hill criterion [4]. According to the criterion:

$$\frac{1}{\sigma_\theta^2} = \frac{\cos^4\theta}{\sigma_1^2} + \sin^2\theta\cos^2\theta\left(\frac{1}{S^2} - \frac{1}{\sigma_1^2}\right) + \frac{\sin^4\theta}{\sigma_2^2}, \tag{9}$$

where σ_1 and σ_2 are the tensile strength in the longitudinal and transverse directions, respectively, and S is the shear strength in principal material co-ordinates. Equations 8 and 9 have been shown to be applicable for various long-fibre aligned composites [5, 6]. In case of injection-moulded, short glass fibre-reinforced thermoplastics samples with well-aligned fibres, Ramsteiner and Theysohn [2] and Ramsteiner [7] have reported good agreement of the experimental data with these equations. In the following we have also used these equations for extruded sheet samples.

3. Experimental procedure

3.1. Sample preparation

Short glass fibre-reinforced polypropylene sheet 12 cm wide and 0.1 cm thick was extruded using Profax PC 072-3 commercial glass fibre-filled polypropylene granules containing 30% by weight of glass fibres. A sheet was also extruded from the base homopolymer under similar conditions.

3.1.1. Fibre-length distribution measurement

A small piece of the composite sample was subjected to a temperature of 500° C in an oven to burn off the polypropylene matrix and the lengths of 700 fibres collected from the sample were measured on a Projectina Microscope from which histogram of fibre-length distribution was prepared. The average fibre radius was determined to be 7 μm.

3.1.2. Mechanical testing

Dumb-bell shaped tensile specimens of the standard size were cut from the extruded sheet and

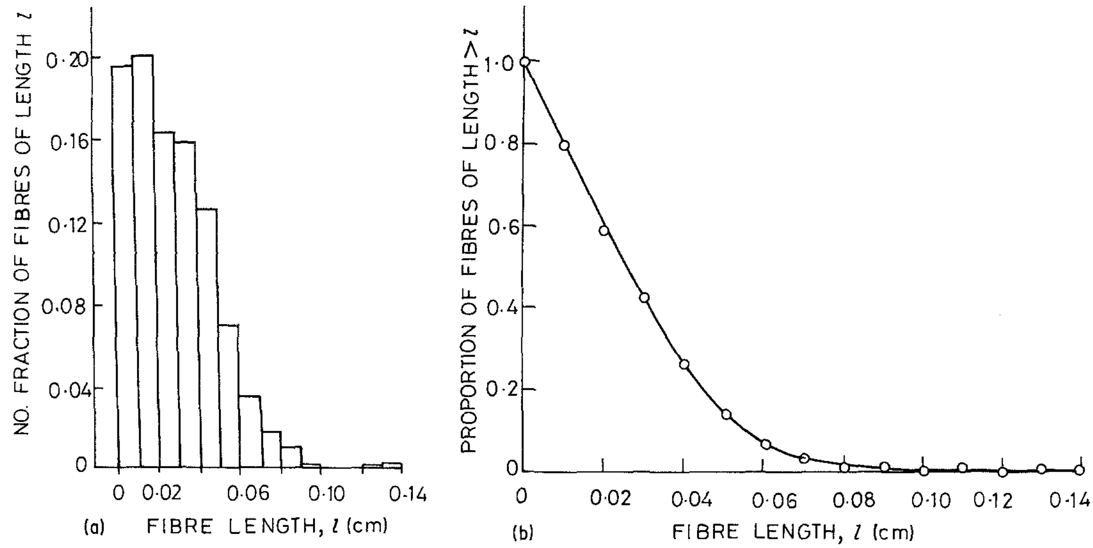

Figure 1 (a) Histogram of fibre length. (b) Fibre-length distribution.

tested on an Instron tensile tester at a strain rate of 1% min^{-1} at $25 \pm 2°$ C. The elastic modulus, tensile strength and elongation-to-break were obtained from the load–elongation curve. In each case, at least five samples were tested and the results were found to be very reproducible.

The torsional modulus of the samples was determined on a torsional tester described in detail elsewhere [8].

4. Results and discussion

The fibre-length distribution data are shown in Fig. 1. The stress–strain curves for the composite samples having various orientations to the load axis and for the base polymer are shown in Fig. 2. The elastic modulus and tensile strength values obtained from these curves are shown in Table I. An attempt to analyse these data in terms of the Bowyer and Bader theory showed that for a particular value of τ, the orientation factor was constant only in the higher strain region [9]. This suggested that the assumption that τ was constant at all strains needed re-examination and this is done here.

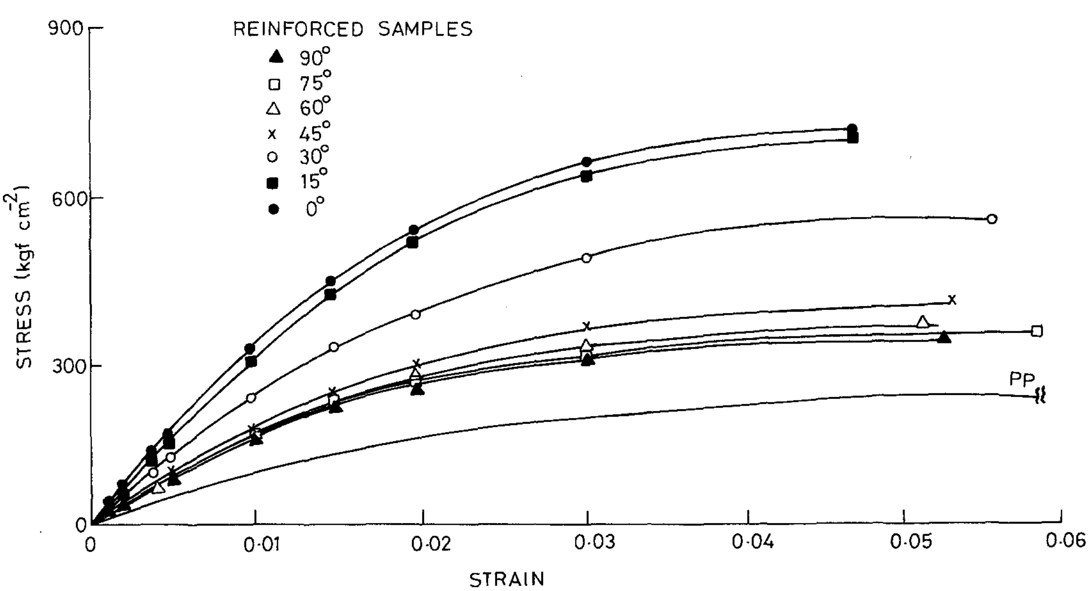

Figure 2 Stress–strain curves for the unreinforced and reinforced sample.

TABLE I Mechanical data on unreinforced and reinforced samples

Sample	Elastic modulus ($kg\ cm^{-2}$)	Tensile strength ($kg\ cm^{-2}$)
Polypropylene	8 200	287
Reinforced polypropylene		
0°	31 400	684
15°	30 750	680
30°	23 700	547
45°	18 070	430
60°	15 900	375
75°	16 600	355
90°	16 800	350

4.1. Fibre alignment and its influence on mechanical properties

The values of elastic modulus and tensile strength at various fibre orientations were analysed according to Equation 8 and 9, respectively, to establish whether or not the fibres are well aligned in the extruded sheet. The measured values of the tensile modulus for various fibre orientations and the best-fit curve based on Equation 8 are shown in Fig. 3. Similarly, Fig. 4 shows the tensile strength values and the best fit curve using Equation 9. From the comparisons shown in Figs 3 and 4 it is seen that the orientation dependence of both stiffness and strength is as expected for aligned fibre composites. The effect of any fibre misalignment or error in preparing the sample so that the fibres have a desired inclination to the true 0° direction is most prominent for the $\theta = 0°$ case because of the predominance of the $\cos\theta$ terms. From the figures, the average misorientation was estimated to be only $\pm 5°$. The theoretical curves in Figs 3 and 4 correspond to the following values of the elastic and strength constants:

$$E_1 = 33\,000\ \text{kgf cm}^{-2}, \qquad \sigma_1 = 720\ \text{kgf cm}^{-2}$$

$$E_2 = 16\,800\ \text{kgf cm}^{-2}, \qquad \sigma_2 = 350\ \text{kgf cm}^{-2}$$

$$\left(\frac{1}{G_{12}} - \frac{2\nu_{12}}{E_1}\right) = 0.132 \times 10^{-3}\ \text{cm}^2\ \text{kgf}^{-1},$$

$$S = 272.5\ \text{kgf cm}^{-2}.$$

It may be noted that the values of E_1, E_2, σ_1 and σ_2 are quite close to the measured values of these parameters given in Table I. As regards G_{12}, the shear modulus; ν_{12}, the Poisson's ratio and S, the shear strength, the following comments may

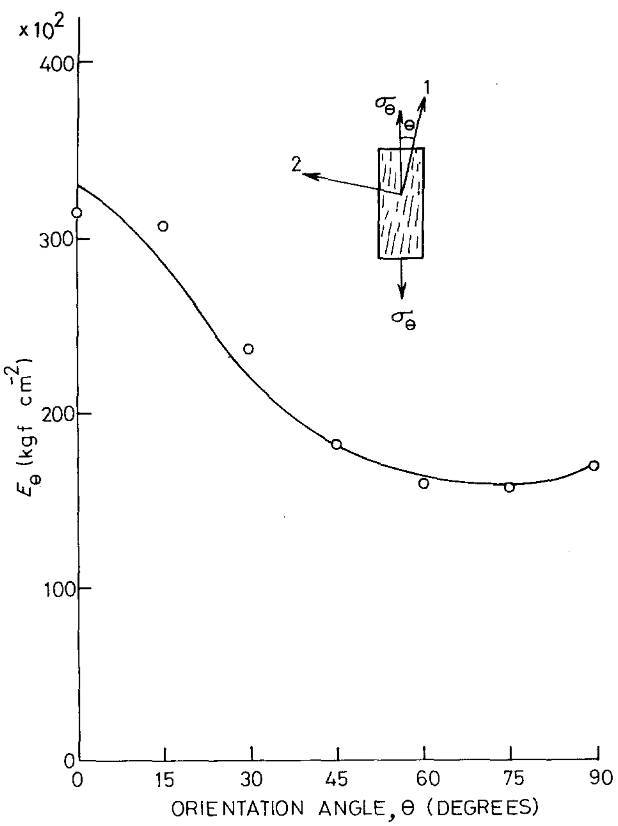

Figure 3 Directional dependence of tensile modulus (o, experimental points; ——, predicted curve).

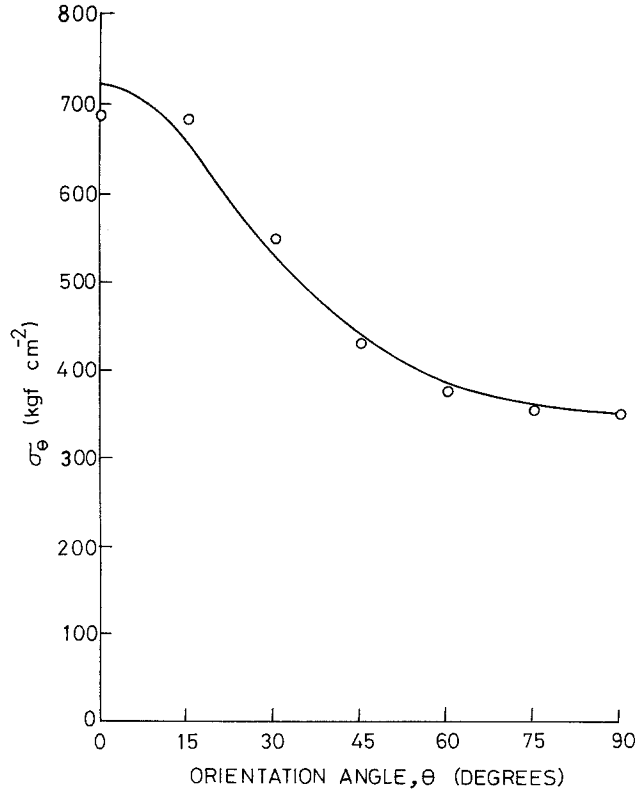

Figure 4 Directional dependence of tensile strength (o, experimental points; ——, predicted curve).

be made. G_{12} comes out to be about 6580 kgf cm^{-2} for $\nu_{12} = 0.33$ and 8403 kgf cm^{-2} for $\nu_{12} = -0.22$. No direct method for the measurement of G_{12} using a prismatic sample is available [4]. A low frequency (1 Hz) torsion pendulum test on a narrow strip sample with fibres oriented along the length gave an approximate value for G_{12} around 8000 kgf cm^{-2}. ν_{12} was not determined. While the shear strength of these samples were not measured, an injection-moulded sample having much poorer fibre orientation was found to have a buckling shear stress on a constant rate of loading type of torsion tester [14] close to 205 kgf cm^{-2}. It would thus appear the macro-mechanical approach is reasonably successful for these samples.

4.2. Determination of the interfacial bond strength using Bowyer and Bader's procedure

The stress–strain curve for the 0°-sample ($\eta \approx 1$) was analysed to obtained the interfacial bond strength, i.e. τ at failure. However, before undertaking this analysis, Equations 4 to 7 were rewritten in the following form:

$$\sigma_c - V_m(\sigma_m)_{\epsilon_c} = \alpha\tau + \beta\epsilon_c - \frac{E_f^2 r_f}{2\tau}(\epsilon_c)^2, \quad (10)$$

where

$$\alpha = \Sigma_{(\text{subcritical})} \frac{L_i V_i}{2r_f}, \quad (11)$$

$$\beta = E_f V_{sup}, \quad (12)$$

and

$$\gamma = \Sigma_{(\text{supercritical})} \frac{V_i}{L_j}. \quad (13)$$

All these symbols have been explained in Section 2 except V_{sup} which is the total volume fraction of supercritical fibres and $(\sigma_m)_{\epsilon_c}$ which denotes the matrix stress corresponding to the composite strain. Also, whereas Bowyer and Bader assumed a linear elastic behaviour for the matrix for all strains (see Equation 7), we use the actual stress–strain curve for the matrix.

From Equations 11 to 13, it is seen that α, β and γ are functions of l_c and the fibre-length distribution only because l_c determines if a fibre is supercritical or subcritical. Using the histogram of fibre length, α, β and γ have been plotted in Fig. 5 as functions of l_c. These graphs simplify the calculations very much.

As suggested by Bowyer and Bader, trial values of τ are considered and using Equations 1 and 4 to 7, η is calculated at several values of ϵ_c. The value of τ, which yields the same value

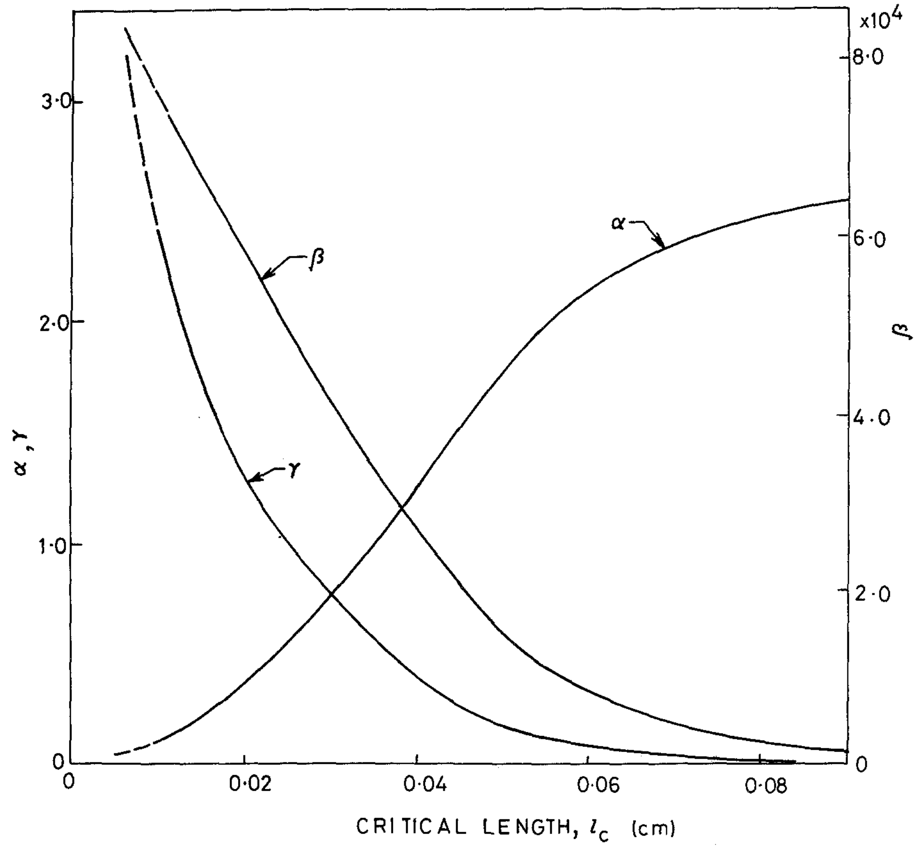

Figure 5 α, β and γ as functions of critical length (see text for details).

of η at all composite strains is selected as the shear strength of the interfacial bond. Since our sample was well characterized with the inclination of the fibres to the load axis very close to 0°, i.e. $\eta \approx 1$, the criterion adopted for the acceptance of τ was that the quotient

$$\frac{\sigma_c - V_m(\sigma_m)_{\epsilon_c}}{X+Y} \approx 1, \tag{14}$$

for all strains. Various values of τ in the range 160 to 240 kgf cm^{-2} were tried but only $\tau =$ 220 kgf cm^{-2} showed any reasonable agreement with Equation 14 and this agreement was limited to the strains close to failure only. Over the entire range of strains considered (0.005 to 0.045), the value of the quotient varied from 0.41 to 0.92. The graphical representation of the results will be given in the next subsection.

The large variation of the quotient (or η) over the range of the composite strains shows that the assumption of the constancy of τ at all strain levels is not valid. From a physical point of view, such an assumption implies that the interface is stressed to its maximum attainable strength even at low composite strains.

Deviating from this assumption, the following two alternatives were considered:

(a) τ is proportional to ϵ_c; and

(b) τ is proportional to σ_c.

The first alternative showed a much wider variation than in the $\tau =$ constant case. So this hypothesis was discarded. However, case (b) implying $\tau = K\sigma_c$, where K is constant, yielded much better results and is discussed below.

4.3. Interfacial shear strength using Equation $\tau = K\sigma_c$

Various trial values of K were considered for the same data. For $K = 0.28$, the Criterion 14 was satisfied quite well over the entire range of strain. This indicates that there is a good agreement between the calculated values of $(X + Y)$ and $(\sigma_c - V_m(\sigma_m)_{\epsilon_c})$ values obtained from the stress–strain curves of the reinforced and the unreinforced samples.

The results of both analyses are compared in Figs 6 to 8. The results labelled "Bowyer and

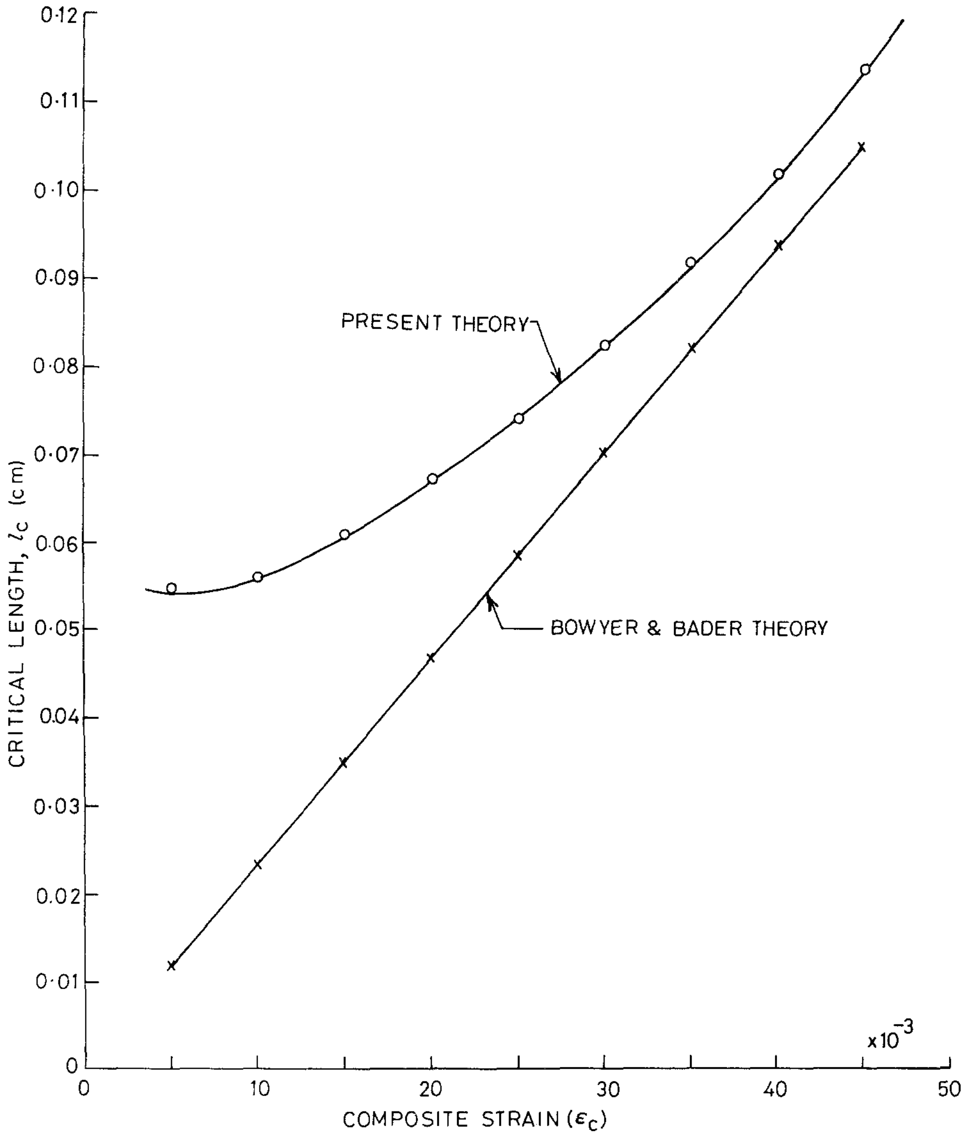

Figure 6 The critical length as a function of composite strain obtained from the two theories.

Bader theory" correspond to the calculations for $\tau = 220\,\text{kgf cm}^{-2}$ in Equation 2, and the results labelled "present theory" are those obtained for $K = 0.28$. It is seen that the values of the critical length (l_c), contribution of subcritical fibres X and that of supercritical fibres Y differ significantly for these two calculations. The comparison between $X + Y$ and $\sigma_c - V_m(\sigma_m)_{\epsilon_c}$ values for both cases is shown in Fig. 8 and the close agreement obtained for the $\tau = K\sigma_c$ case (present theory) is noteworthy and discussed below. In Fig. 8 the quotient η, as defined in Equation 14, has also been plotted for both calculations. The "present theory" values over the entire strain range are close to $\cos^4(5°) = 0.984$ where 5° was found to be the average misorientation (see Section 4.1 above) and the expression $\eta = \cos^4\theta$ was suggested by Bowyer and Bader [1] for aligned fibre composites. However, the Bowyer and Bader theory values of η approach 0.984 only near the failure strain.

The experimental results suggest very strongly that the shear stress acting at the interface between the fibre and the matrix is a function of the composite stress σ_c for a well-aligned short fibre-reinforced polypropylene sample loaded in tension along the fibre direction. Moreover, this functional relationship is linear or very

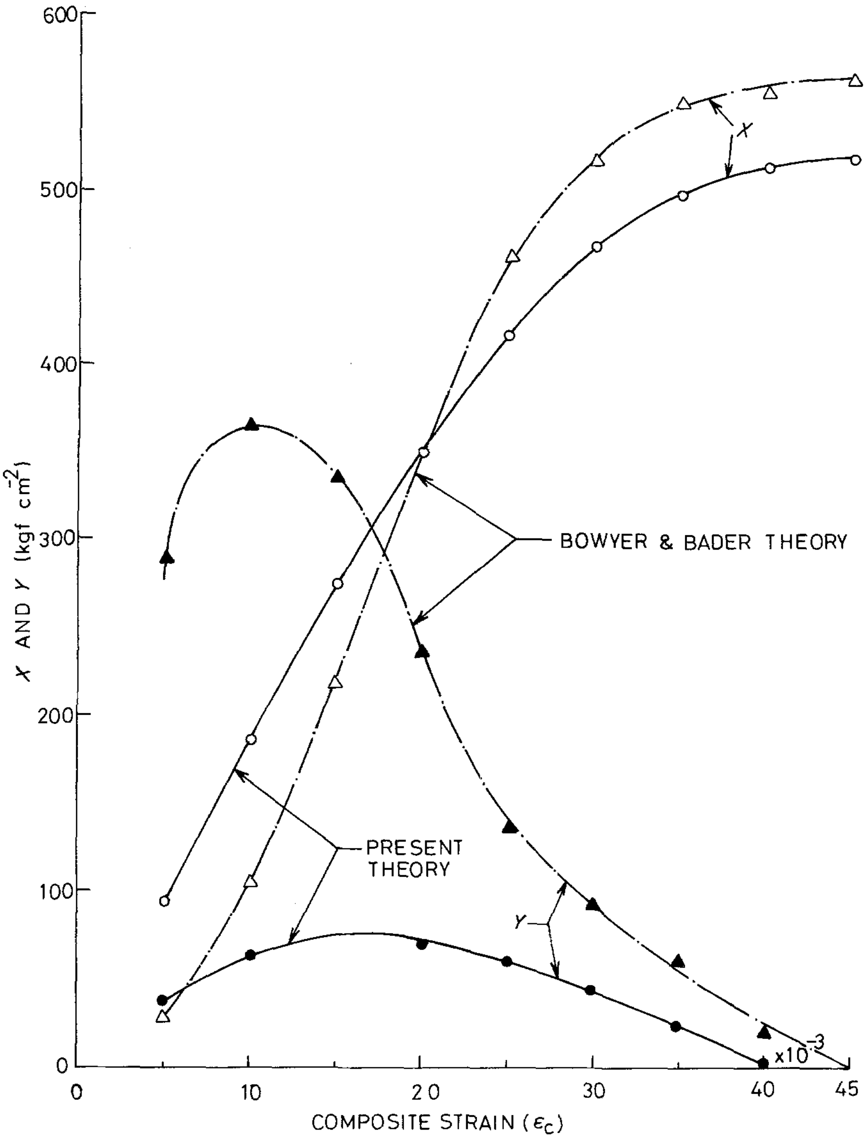

Figure 7 The parameters X and Y as functions of composite strain obtained from the two theories.

nearly so. Various theoretical models for the same problem, assuming both fibre and matrix to be linearly elastic, show that the interfacial shear stress is proportional to the applied load. These models have been discussed in various monographs (see, for example [10, 11]). A recent paper [12] addressed to this problem uses shear-lag analysis. That this proportionality is nearly true for a non-linear, non-elastic matrix material in the range of strains considered (0.005–0.045) is an interesting result. Outwater [13] has proposed a model, specifically for aligned fibre-reinforced polymers, which attributes the interfacial shear strength to friction between the fibre and the matrix caused by shrinkage stresses generated during curing or solidification of the matrix. In that sense, K plays the role of the coefficient of friction.

Finally, the value of the interfacial bond strength, i.e. the value of τ at failure, has been found to be 204.1 kgf cm^{-2}. Ramsteiner and Theysohn found this value to be 225 kgf cm^{-2} for the composites using chemically modified polypropylene. Also our value compares favourably with the predicted value of the matrix shear strength. The tensile strength of the unreinforced polypropylene was found to be 287 kgf cm^{-2}. From the equivalence of tensile stress and shear

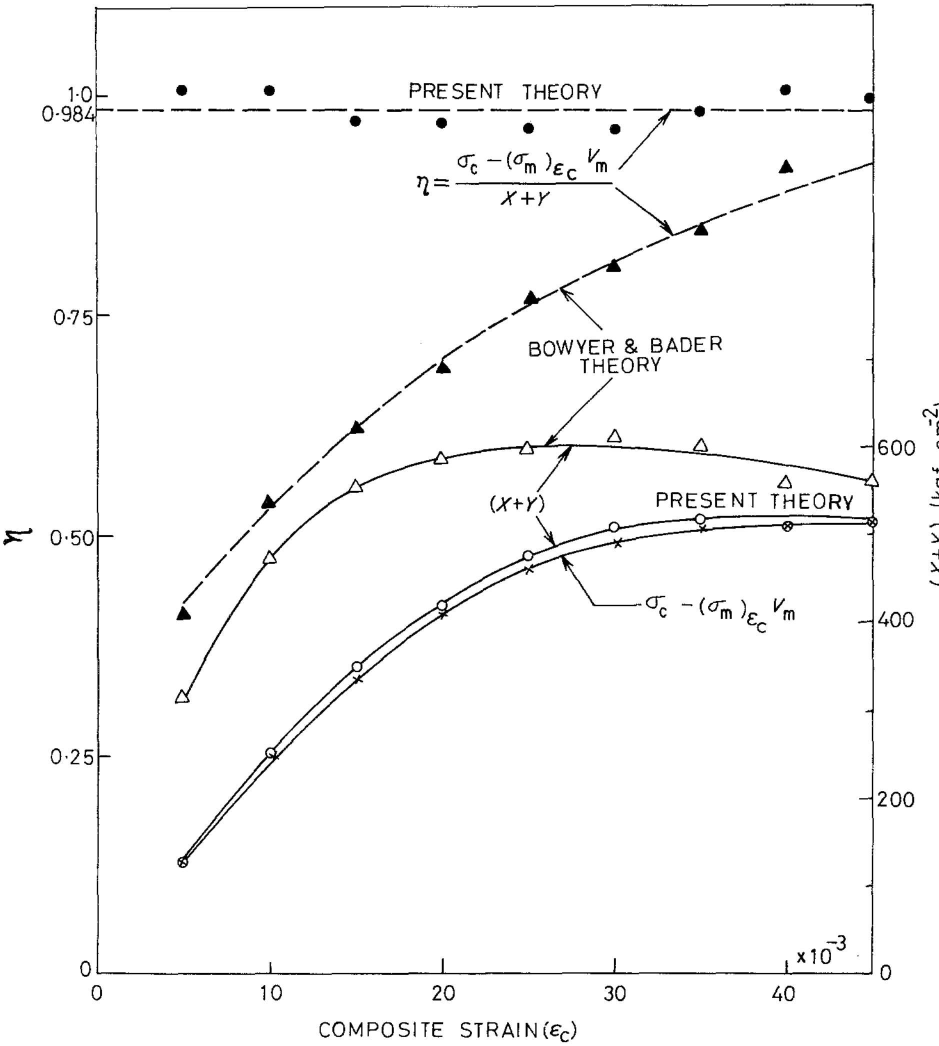

Figure 8 The orientation factor, η as calculated from Bowyer and Bader theory and the present theory.

stress, i.e. $\tau = \sigma/\sqrt{2}$ for polypropylene [14], the shear strength of the matrix material is found to be 202 kgf cm^{-2}.

5. Conclusions

It has been shown in this paper that the use of the linear relationship between the interfacial shear stress and the composite stress, i.e. $\tau = K\sigma_c$, produces a close agreement between $[\sigma_c - V_m(\sigma_m)_{\epsilon_c}]$ and $[X + Y]$. Since X and Y, respectively, denote the average stresses to which the subcritical and supercritical fibres are subjected, the values of X and Y will be in error if l_c is not correctly determined. Thus, in our view, the principal contribution of the equation $\tau = K\sigma_c$ is the proper evaluation of the critical length during the deformation process. Equation 1 can now be written as:

$$l_c = \frac{E_f r_f \epsilon_c}{K\sigma_c} = \left(\frac{E_f r_f}{K}\right)\left(\frac{\epsilon_c}{\sigma_c}\right). \quad (15)$$

Hence the critical length is inversely proportional to the secant modulus of the stress–strain curve of well-aligned short glass fibre-reinforced polypropylene sample loaded in tension in the direction of fibres. On the other hand, according to Bowyer and Bader's procedure, l_c is proportional to ϵ_c since τ is constant.

Secondly, for the purpose of determining the interfacial bond strength, it is preferable to use the 0° fibre orientation stress–strain curve because there is no component of the applied stress acting transverse to the fibres. Intuitively it is expected that this transverse component will influence the value of K. This point is under consideration. Another aspect of the effect of fibre orientation (θ) deals with the stiffness and strength properties of the composite. It has been shown that Equations 8 and 9 described this effect adequately for short fibre-reinforced thermoplastics. The use of a single orientation factor for both stiffness and strength of composites, as adopted by many workers in this area, is erroneous since, from the mathematical point of view, it is equivalent to replacing the fourth order and second order tensor transformation relations by a single scalar factor.

Finally, comparing our procedure for the determination of the interfacial bond strength with others as reported above, it is stated that our procedure is consistent and economical since the stress–strain curve of only one volume fraction and one orientation of fibres is required.

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