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Theoretical modelling of tensile properties of short sisal fibre-reinforced low-density polyethylene composites

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The experimentally observed tensile properties (tensile strength and modulus) of short sisal fibre-reinforced LDPE with different fibre loading have been compared with the existing theories of reinforcement. The macroscopic behaviour of fibre-filled composites is affected by fibre loading, orientation and length of the fibres in the continuous medium. The interfacial adhesion between fibre and matrix also plays a major role in controlling the mechanical properties of the fibre-filled composites. In this study, a comparison is made between experimental data and different theoretical models. Composite models, such as parallel and series, Hirsch, Cox, Halpin–Tsai, modified Halpin–Tsai and modified Bowyer and Bader, have been tried to fit the experimental data.

1. Introduction

The mechanical properties of fibre-filled composites are affected by a number of parameters such as fibre length, fibre orientation, fibre dispersion, fibre geometry and the degree of interfacial adhesion between fibre and matrix [1–5]. In the literature, a number of equations and theories have been developed to describe the relation between these parameters and properties of constituent components of composites. The efficiency of load transfer from matrix to fibre in a composite is strongly related to the optimum mechanical properties of the composite.

One of the earliest theories of reinforcement developed by Cox [6] is based on shear-lag mechanism observed in fibrous composites. According to Cox, in shear-lag analysis, the main aspects of controlling the properties of a composite are critical length of the fibre and interfacial shear strength between fibre and matrix. The critical length of the fibre, l_c, in composites is a parameter which determines the amount of stress transferred to the fibre. That is, if the length-to-diameter ratio is higher than the critical aspect ratio, composites show superior properties, while for a fibre whose aspect ratio is smaller than the critical aspect ratio, composites show weaker properties. In Cox's treatment, interfacial shear strength is produced on the surface of the fibre due to the "shear lag" between fibre and matrix during the failure of the composite. However, Cox's shear-lag analysis has two major disadvantages. The first one is that stress amplification effects at the fibre ends are not taken into account, and the second is that the matrix tensile stress possesses no radial dependence.

Piggot modified Cox's theory by introducing a new theory which combines plastic deformation at the fibre ends with elastic deformation towards the centre of the fibre during tensile loading [7].

It was also reported that the strength of short fibre-reinforced thermoplastics and thermosets are highly dependent on two factors, such as "fibre orientation factor" and "fibre length factor" which contribute to the strength of the composite [8]. In another study, Piggot developed an equation for calculating the minimum volume fraction of fibres for a good reinforcement [9].

Termonia [10] presented a computer model to study the effect of fibre characteristics on the mechanical properties of short fibre-reinforced composites. Their studies revealed that the effect of fibre orientation on modulus and tensile strength of the composite is very weak and an optimum fibre aspect ratio is essential for effective reinforcement. They modelled the whole composite material as a three-dimensional lattice of bonds having different elastic constants for the fibre and for the matrix. They also found that in composites, a micro-failure mechanism originates at the fibre ends and propagates along the fibre–matrix interface with no fibre breaking. Termonia [10] calculated a value for the reinforcement efficiency factor based on their observations in that study.

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Monette *et al.* [11] suggested a computer modelling for the theoretical understanding of the concept of critical length in composites. The effects of interface and matrix properties on critical aspect ratio was also studied by them. They found that critical aspect ratio is related to interfacial shear strength and fibre strength only and not to matrix properties. They further showed that under certain circumstances, matrix viscosity and strain rate can influence the critical aspect ratio. Karam [12] studied the effect of fibre volume fraction on the strength properties of short fibre-reinforced cement. He proposed a modification to the existing model in order to calculate the strength of the composites. The modification is based on the reduction of interfacial surfaces due to fibre–fibre and fibre–void interactions.

2. Theory

Several theories have been proposed to model the tensile properties of composite material in terms of different parameters [13–16]. These theories can be classified into two groups, based on the nature of the matrix and the reinforcements. These are the theories of reinforcements in a non-rigid matrix and those in a rigid matrix.

2.1. Theories of rigid particulate reinforcement in non-rigid polymer matrices

2.1.1. Einstein and Guth equations

These equations are mainly used for the theoretical calculation of the properties of particulate (spherical)-reinforced polymer composites [17, 18].

According to the Einstein equation

$$M_c = M_m(1 + 1.25V_p) \quad (1)$$

where M_c and M_m are the Young's modulus of composite and matrix, respectively, V_p is the particle volume fraction. Guth derived an equation [18]

$$M_c = M_m(1 + 2.5V_p + 14.1V_p^2) \quad (2)$$

This equation is further related to the tensile strength, T_c, of the composite

$$T_c = T_m(1 - V_p^{2/3}) \quad (3)$$

where T_m is the tensile strength of the matrix.

2.1.2. Modified Guth equation

Cohan [19] introduced a particle shape factor, *S*, for non-spherical particles, where *S* is defined as the ratio of the length-to-width of particles.

The modified Guth relation is given by

$$M_c = M_m(1 + 0.675\,SV_p + 1.62\,S^2V_p^2) \quad (4)$$

2.1.3. Modified Kerner equation

Young's modulus of spherically shaped particulate-filled polymer composites is given by Kerner's equation [20]

$$M_c = M_m\left[1 + \frac{V_p 15(1 - \sigma_m)}{V_m(8 - 10\sigma_m)}\right] \quad (5)$$

where V_m is the matrix volume fraction, σ_m is the Poisson's ratio of the matrix.

Nielsen modified Kerner's equation by introducing a function called the particle packing factor, *BF* [21]

$$M_c = M_m\left(\frac{1 + AB\,V_p}{1 - BF\,V_p}\right) \quad (6)$$

A accounts for the factors such as geometry of the filler and Poisson's ratio of the matrix. *B* accounts for the relative moduli of filler and matrix

$$B = \frac{M_p/M_m - 1}{M_p/M_m + A} \quad (7)$$

where M_p and M_m are the Young's moduli of particulate filler and matrix, respectively.

Equations 1–7 are mainly applicable only in the field of particulate-reinforced polymers, especially in non-rigid polymer matrices.

2.2. The theories of rigid reinforcement (particulate and fibrous) in a rigid matrix

These theories can be successfully applied in the systems of both particulate and fibrous reinforcement.

2.2.1. Parallel and series model

According to these models, Young's modulus and tensile strength are calculated using the following equations.

Parallel model

$$M_c = M_f V_f + M_m V_m \quad (8)$$

$$T_c = T_f V_f + T_m V_m \quad (9)$$

Series model

$$M_c = \frac{M_m M_f}{M_m V_f + M_f V_m} \quad (10)$$

$$T_c = \frac{T_m T_f}{T_m V_f + T_f V_m} \quad (11)$$

where M_c, M_m and M_f are the Young's moduli of composite, matrix and fibre, respectively. T_c, T_m and T_f are the tensile strength of the composite, matrix and fibre, respectively.

In the case of a parallel model, it is assumed that isostrain conditions exist for both matrix and fibre, whereas in the case of a series model, stress was assumed to be uniform in both matrix and fibre [22].

2.2.2. Hirsch's model

Hirsch's model is a combination of parallel and series models [23]. This model can be represented by Fig. 1.

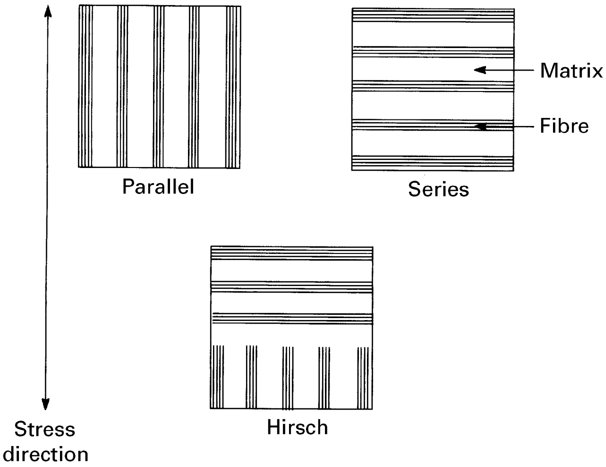

Figure 1 A schematic representation of Hirsch's model.

According to this model, Young's modulus and tensile strength are calculated using the following equations

$$M_c = x(M_m V_m + M_f V_f) + (1 - x)\frac{M_f M_m}{M_m V_f + M_f V_m} \quad (12)$$

$$T_c = x(T_m V_m + T_f V_f) + (1 - x)\frac{T_f T_m}{T_m V_f + T_f V_m} \quad (13)$$

The parameter, x, is explained in Section 4.

2.2.3. The Halpin–Tsai model

This model has been used by several researchers in the system of polymeric blends which consist of continuous and discontinuous phases [24, 25]. However, it was reported that this model was also useful in determining the properties of composites that contain discontinuous fibres oriented in the loading direction [26–28].

According to Halpin–Tsai, Young's modulus, M_c, of the composite is given by

$$M_c = M_m\left(\frac{1 + A\eta V_f}{1 - \eta V_f}\right) \quad (14)$$

$$T_c = T_m\left(\frac{1 + A\eta V_f}{1 - \eta V_f}\right) \quad (15)$$

$$\eta = \frac{M_f/M_m - 1}{M_f/M_m + A} \quad (16)$$

$$\eta = \frac{T_f/T_m - 1}{T_f/T_m + A} \quad (17)$$

where A is the measure of fibre geometry, fibre distribution and fibre loading conditions.

2.2.4. Modified Halpin–Tsai equation

Nielson modified the Halpin–Tsai equation by including the maximum packing fraction, ϕ_{max}, of the reinforcement [29]. According to this

$$M_c = M_m\left(\frac{1 + A\eta V_f}{1 - \eta\psi V_f}\right) \quad (18)$$

$$T_c = T_m\left(\frac{1 + A\eta V_f}{1 - \eta\psi V_f}\right) \quad (19)$$

$$\psi = 1 + \left(\frac{1 - \phi_{max}}{\phi_{max}^2}\right)V_f \quad (20)$$

$$A = K - 1 \quad (21)$$

$$K = 1 + 2l/d \quad (22)$$

η is given by Equations 16 and 17, and accounts for the relative moduli of fibre and matrix, respectively. ψ depends upon the particle packing fraction. The value of A is determined from the Einstein coefficient, K, reported in a previous study [30]. ϕ_{max} is the maximum packing fraction, and has a value 0.785 for square arrangement of fibres, 0.907 for hexagonal array of fibres and 0.82 for random packing of fibres.

2.2.5. Cox model

According to Cox's theory, longitudinal Young's modulus, M_c, is given by the equation [6]

$$M_c = M_f V_f\left(1 - \frac{\tan h\beta 1/2}{\beta 1/2}\right) + M_m V_m \quad (23)$$

where M_m and M_f are the Young's moduli of matrix and fibre

$$\beta = \left[\frac{2\pi G_m}{M_f A_f \ln(R/r)}\right]^{\frac{1}{2}} \quad (24)$$

where r is the radius of the fibre, G_m the shear modulus of matrix, R the centre-to-centre distance of the fibres, and A_f the area of the fibre.

For hexagonally packed fibres

$$R = \left(\frac{2\pi r^2}{3^{1/2} V_f}\right)^{\frac{1}{2}} \quad (25)$$

For square packed fibres

$$R = r\left(\frac{\pi}{4V_f}\right)^{\frac{1}{2}} \quad (26)$$

According to Cox's model, tensile strength, T_c, is given by [6]

$$T_c = T_f V_f\left(1 - \frac{\tan h\beta 1/2}{\beta 1/2}\right) + T_m V_m \quad (27)$$

β is given by the Equation 24.

2.2.6. Modified Bowyer and Bader's model

According to Bowyer and Bader's model, the tensile strength of short fibre-reinforced thermoplastic composites is the sum of contributions from subcritical and supercritical fibres and that from the matrix [31]. Tensile strength is given by

$$T_c = T_f K_1 K_2 V_f + T_m V_m \quad (28)$$

where K_1 is the fibre orientation factor. Depending on fibre orientations, K_1 also changes [8]. K_2 is the fibre length factor.

For fibres with $l > l_c$.

$$K_2 = l - l_c/2l \quad (29)$$

TABLE I Physical and mechanical properties of low-density polyethylene (LDPE-Indothene 16MA 400)

Melt flow index (g/10 min.)	Density (g cm^{-3})	Tensile strength at break (MPa)	Elongation at break (%)	Modulus of elasticity (MPa)	Vicat softening point (°C)	Crystalline melting point (°C)
40	0.916	9	200	140	85	104

For fibres with $l < l_c$.

$$K_2 = l/2l_c \tag{30}$$

where l is the length of the fibre and l_c is the critical length of the fibre. Young's modulus also can be calculated using the same equation

$$M_c = M_f K_1 K_2 V_f + M_m V_m \tag{31}$$

The main purpose of this study was to correlate the experimental tensile properties of short sisal fibre-reinforced low-density polyethylene with theoretical values, calculated by various theoretical models. It was found that some of the models show a good agreement with experimental values. The models clearly indicate that the parameters such as fibre orientation, fibre length, fibre loading, fibre dispersion and interfacial shear strength between fibre and matrix play a major role in contributing to the tensile properties of short fibre-reinforced polymer composites. The limitation of these models was also considered in this study.

3. Experimental procedure

Sisal fibre was obtained from local sources. LDPE, graded as Indothene 16 MA 400, was supplied by Indian Petrochemical Corporation Ltd, Baroda, India. The properties of sisal and LDPE are listed in Tables I and II.

Sisal/LDPE composites were prepared by the solution mixing technique. Fibre was added to a viscous slurry of polyethylene in toluene which was prepared by adding toluene to a melt of the polymer. The mixing was carried out manually in a stainless steel beaker using a stainless steel stirrer. The temperature was maintained at 110 °C during mixing for about 10 min. The mix was then transferred into a flat tray as lumps and kept in a vacuum oven at 70 °C for 2 h to remove the solvent. Composites containing 10%, 20%, and 30% by weight of fibre were prepared using fibres of length in the range 2–10 mm. The mix is then extruded through a ram-type hand-operated injection-moulding machine at a temperature of 125 ± 3 °C. For the preparation of longitudinally and transversally oriented composites, the extrudates having a diameter of 4 mm were collected and aligned in a leaky mould [32]. They were then compression moulded at pressure of about 8 MPa and at a temperature of 125 ± 3 °C. The composites so obtained were removed after cooling the mould below 50 °C. Rectangular specimens of size 120 mm × 20.5 mm × 2.5 mm were cut from above composites for further testing. Randomly oriented composite sheets (120 mm × 12.5 mm × 3 mm) were prepared by standard injection moulding of the mix using a ram-type hand-injection moulding machine. Fig. 2. shows the optical photographs of different orientations of the fibre in the matrix. Tensile testing of the composites was carried out using an Instron Universal Testing Machine model 1190 at a crosshead speed of 200 mm min^{-1} and gauge length of 50 mm. At least five specimens were tested for each set of samples and the mean values are reported.

TABLE II Physical and mechanical properties of sisal fibre

Density (g cm^{-3})	Tensile strength (GPa)	Young's modulus (GPa)	Elongation at break (%)	Diameter (μm)
1.41	0.4–0.7	9–20	5–14	100–300

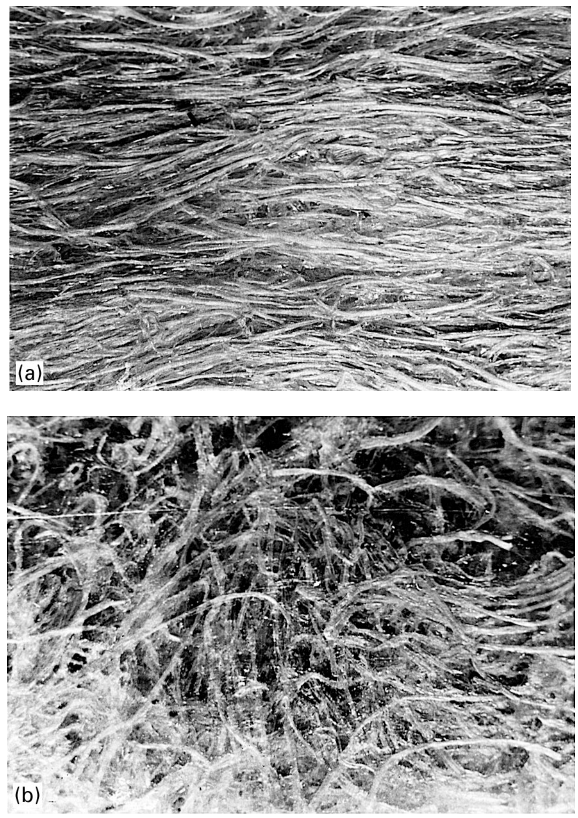

Figure 2 Optical photographs of the composites showing the different orientation of fibres: (a) parallel and (b) random. ×60.

4. Results and discussion

Composites containing 2–10 mm length fibres were analysed in this study. However, it was found that composites with incorporated 6 mm fibre lengths showed maximum tensile strength [33]. Therefore, 6 mm fibre length was used in this study. Fig. 3 shows a comparison of the variation in theoretical and experimental tensile strength values of longitudinally oriented composites with volume fraction of fibres.

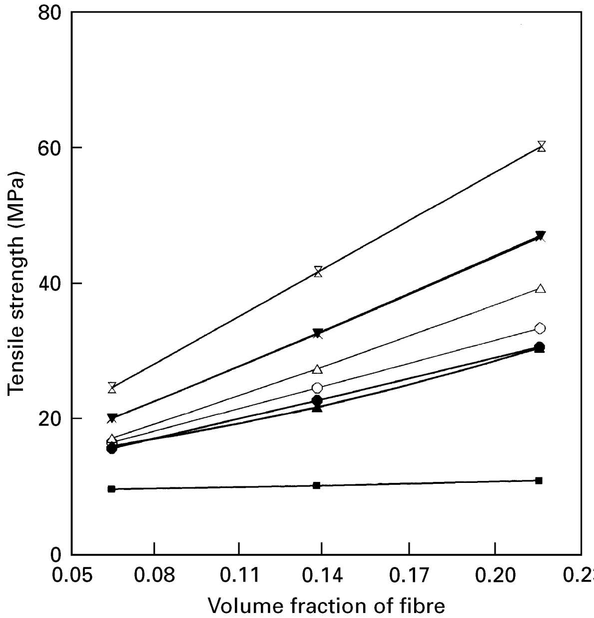

Figure 3 Variation of experimental and theoretical tensile strength values of longitudinally oriented composites as a function of volume fraction of fibres: (▲) Experimental, (⧖) parallel, (■) series, (⬣) Hirsch, (×) Halpin–Tsai, (▼) modified H–T, (△) Cox, (⬡) modified Bowyer and Bader.

Theoretical values were calculated using the various models as shown in the figure. It can be seen that, in all cases, tensile strength increases regularly with increase in the volume fraction of fibres. A good correlation between the theoretically and experimentally observed tensile strength was seen in those models predicted using Hirsch and modified Bowyer and Bader equations. The curves showing parallel and series models agree the least with the experimental values. Usually, parallel and series models are used to describe the strength of continuous fibre-reinforced ploymeric composites. The assumption of either uniform stress or uniform strain is clearly an over simplification in this case. The stress-transfer mechanism of continuous fibre-reinforced composite is different from that of short-fibre composites. In the case of short-fibre composites, the stress transfer depends largely on fibre orientation, stress concentration at the fibre ends, critical fibre length, etc. It can be seen from Fig. 1 that, at low volume fraction of the fibres, series and parallel models show a marginal agreement with experimental values. This can be attributed to the fact that, at low volume fraction of the fibre, uniform stress or strain in the composite is achieved as a result of the better distribution of load through the well-dispersed fibres in the matrix. But at high volume fraction, some of the fibre will be agglomerated in the matrix. Hence the applied load will be distributed unevenly between non-aggregated and aggregated fibres.

The Hirsch model is, in fact, a combination of parallel and series models. The agreement between theoretical and experimental values has been found only when the value of x in Equation 13 is 0.4 for longitudinally oriented composites. From this equation, it was found that x is a parameter which determines the stress transfer between fibre and matrix. It is assumed that the controlling factors for the value of x are mainly fibre orientation, fibre length and stress amplification effect at the fibre ends. Thus it is seen that the value of x is a determining factor in describing the real behaviour of short-fibre composites.

The modified Bowyer and Bader model as given in Equation 28, deals with two factors, such as fibre orientation factor, K_1, and fibre length factor, K_2. It was reported that the value of K_1 is 1 for aligned short fibre-reinforced composites in the applied stress direction [8]. The value of K_2 can be calculated using either Equation 29 or 30, because when $l = l_c$, both equations give the same value. The values of K_1 and K_2 were found to be 1 and 0.5 in good agreement with experimental values.

Cox's model in Fig. 3 is applicable only for the system in which the fibre and matrix remain elastic in their mechanical response, where the fibre–matrix interface is perfect and where no axial load is transmitted through the fibre ends [6]. Cox's model shows a reasonable agreement in tensile strength values especially at low volume fraction of the fibre with the experimental values.

The curves showing the tensile strength values from the Halpin–Tsai (H–T) and the modified H–T equation given by Nielson, are depicted in Fig. 3. It can be seen that in both cases, tensile strength values are approximately the same. This clearly reveals that introduction of a factor which determines the maximum packing fraction of fibres has little effect on tensile strength. In this model, the tensile strength values were calculated using Equation 15. The value ϕ was taken by assuming that fibres are arranged in randomly oriented close packed manner. The values of η, ψ, A and K are calculated using Equations 17, 20, 21 and 22, respectively.

Fig. 4 shows a comparison of the variation in experimental and theoretical Young's modulus of

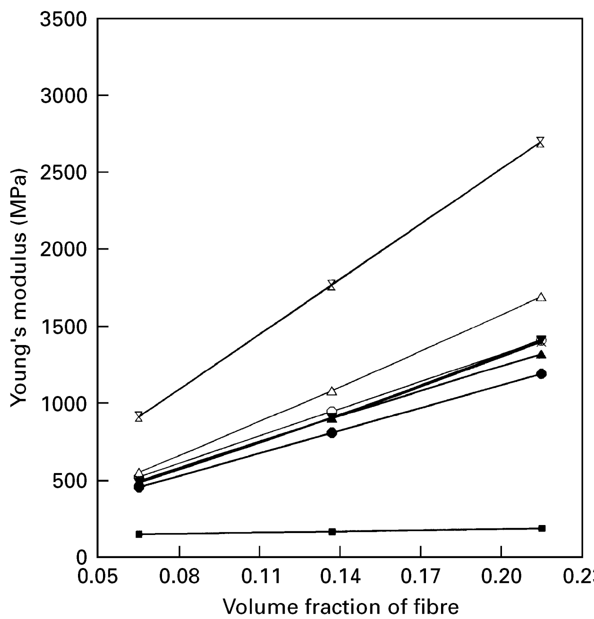

Figure 4 Variation of experimental and theoretical Young's modulus values of longitudinally oriented composites as a function of volume fraction of fibres. For key, see Fig. 3.

longitudinally oriented composites with the volume fraction of fibres. It was observed that a very reasonable correlation exist between theoretical and experimental values in most of the models except series and parallel models. A good agreement is seen in the case of Halpin–Tsai, and modified Bowyer and Bader models. In the case of the Halpin–Tsai and modified H–T models, a good agreement between experimental and theoretical Young's modulus values was observed as compared to the fit of the experimental and theoretical tensile strength values. Fig. 5 shows a comparison of the variation in theoretical as well as experimental Young's modulus values of randomly oriented composites with volume fraction of fibres. The Hirsch and modified Bowyer and Bader models are used for the calculation of theoretical values. Both models partially agree with the experimental values. All other models used in the study are not applicable in the system of randomly oriented composites. In the case of Hirsch's model, tensile strength and Young's modulus values were calculated using Equations 12 and 13. In this case, agreement between experimental and theoretical values has been found only when the value of x in Equations 12 and 13 is 0.1 for randomly oriented composites. The value of x has same meaning as in the case of longitudinally oriented composites. In the case of the modified Bowyer and Bader model, tensile strength and Young's modulus values were calculated using Equations 28 and 31. The value of K_2 in the equation is the same for both longitudinally and randomly oriented composites, but fibre orientation factor, K_1, is different for randomly oriented composites. In this case, the value of K_1, for good agreement between theoretical and experimental values, was found to be 0.2, because it has already been reported that the value of K_1 for fibres arranged in the random fashion is 0.2 [8].

From Figs 3–5, it is clearly observed that, in general, tensile properties of both longitudinally and randomly oriented composites show a reasonable agreement with all the models at low volume fraction of fibres. This may be due to proper orientation of fibres and uniform distribution of applied load as a result of well-dispersed fibres in the matrix at low volume fraction of the fibres.

Fig. 6 shows a comparison of the variation in theoretical and experimental tensile properties of randomly oriented composites as a function of fibre length. The modified Bowyer and Bader model is used for the calculation of tensile properties. The critical length of the fibre was found to be 6 mm from the earlier study [33]. Hence the other lengths of 2 and 10 mm are taken as subcritical and supercritical lengths, respectively. The 2, 6 and 10 mm fibre composites were prepared at 10, 20 and 30 vol% fibres. Tensile properties were calculated using Equations 28 and 31. The value of K_2 in Equations 28 and 31 is different for different fibre lengths. For 2 and 10 mm fibres, the values of K_2 were calculated using Equations 30 and 29, respectively. But any of the above equations can be used for calculating K_2 in the case of 6 mm fibres, because when $l = l_c$ both equations give the same values.

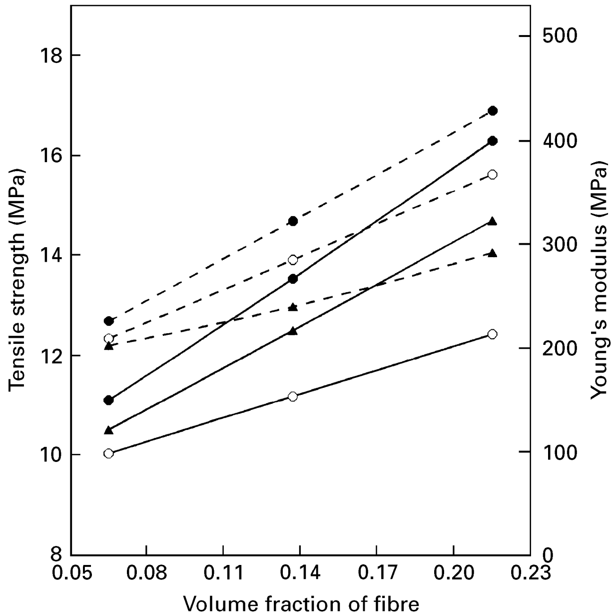

Figure 5 Variation of experimental and theoretical tensile properties of randomly oriented composites as a function of volume fraction of fibres. (——) Strength, (– – –) modulus; (▲) experimental, (⬢) Hirsch, (⬡) modified Bowyer and Bader.

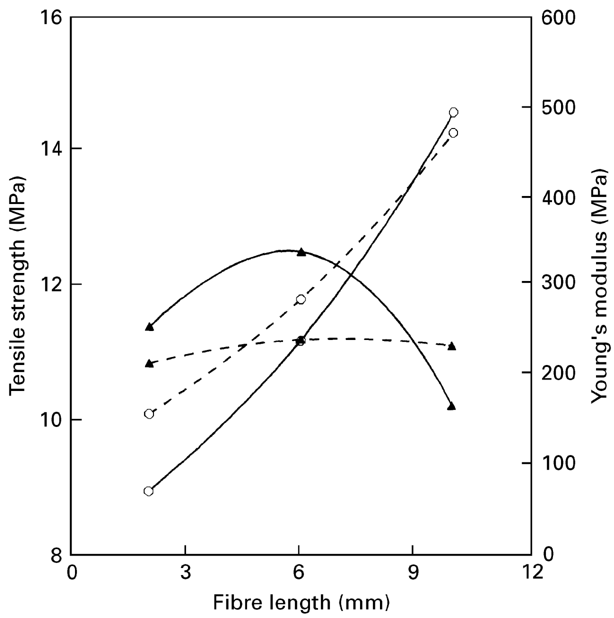

Figure 6 Variation of experimental and theoretical tensile properties of randomly oriented composites as a function of fibre length. For key, see Fig. 5.

It can be seen from Fig. 6 that at 6 mm fibre length, there is a good agreement between theoretical and experimental values in the case of tensile strength and Young's modulus values. This clearly indicates that at critical fibre length, composites show maximum properties.

The limitation of the models used in this study mainly depends on different factors. The chance of the formation of microvoids between fibre and matrix during the preparation of composites greatly influences the tensile properties. This factor is not accounted for in any of the models used in this study. In all models used here, it is assumed that the fibres are

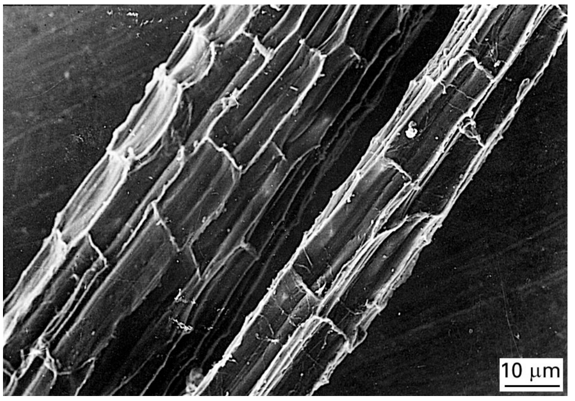

Figure 7 Scanning electron micrograph of the surface of the sisal fibre.

cylindrically shaped. However, the actual shape of the sisal fibre is not perfectly cylindrical due to surface irregularities. Fig. 7 is clear evidence for this fact. The non-uniform shape of the sisal also accounts for the deviation of the tensile properties from the theoretical predictions.

5. Conclusion

A comparison between experimental results and the prediction from theory of the tensile properties (tensile strength and Young's modulus) of short sisal fibre-reinforced low-density polyethylene composites has been presented. The models selected were series and parallel, Hirsch, Halpin–Tsai, modified Halpin–Tsai, Cox, and modified Bowyer and Bader models. Tensile properties of longitudinal and randomly oriented composites were presented as a function of volume fraction of the fibres. All models were applied in the system of longitudinally oriented composites, but the Hirsch and modified Bowyer and Bader equations were applied in the case of randomly oriented composites. All models except the series and parallel model show reasonable agreement with experimental tensile properties of longitudinally oriented composites, especially at low volume fraction of the fibre. Among the various models, the Hirsch and modified Bowyer and Bader equations show very good correlation with experimental results. Hirsch and Bowyer and Bader models also show good correlation with experimental results of randomly oriented composites. The effect of fibre length on tensile properties was also analysed in this study. The Bowyer and Bader model revealed that the critical length plays a major role in contributing to the tensile properties of short fibre-reinforced polymer composites. All theoretical models used in this study clearly indicate that tensile properties of short fibre-reinforced composites strongly depend on fibre length, fibre loading fibre dispersion, fibre orientation and fibre–matrix interfacial bond strength. Parameters concerning the surface irregularities of sisal fibre and microvoids formed between fibre and matrix are not accounted for in this study.

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