

On the re-inforcement of thermoplastics by imperfectly aligned discontinuous fibres

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Studies of deformation behaviour of short fibre reinforced thermoplastics are complicated by the facts that usually a wide range of fibre lengths are present in moulded test pieces and that the fibres are not systematically oriented with respect to any test direction. An equation has been derived for the stress/strain curve of such a material. This has been used to determine fibre/matrix bond strengths in two glass/nylon 6.6 and two glass/polypropylene composites from measured stress/strain curves and fibre length distributions.

It is concluded that major improvements in the properties of these materials will only be achieved by modifying processing to retain longer fibres.

List of symbols

E_c = Modulus of composite
 E_f = Modulus of fibre
 E_m = Modulus of matrix
 V_f = Volume fraction of fibre
 σ_c = Stress in the composite
 σ_f = Peak stress in a fibre
 $\bar{\sigma}_f$ = Average stress in a fibre
 σ_{uf} = Ultimate strength of fibres
 σ_m' = Stress in matrix at fibre failure strain
 σ_{uc} = Ultimate strength of the composite
 ϵ_c = Strain in composite
 ϵ_{uc} = Ultimate strain of the composite
 τ = Shear strength of the fibre matrix bond
 L = Fibre length
 L_e = Critical fibre length at a composite strain ϵ
 L_c = Critical fibre length for fibre failure
 r = Fibre radius

1. Introduction

Composites of very high specific strength and stiffness can be produced by incorporating continuous aligned filaments of very strong materials (e.g. glass and carbon) into matrices of thermosetting plastics such as the epoxy or polyester materials. The incorporation of similar fibres into thermoplastics matrices is only practicable if the melt fabricating qualities of the thermoplastics are retained, and in practice this means that the fibres must be in short lengths and

present only in moderate amounts. Stiffness and tensile strength may be increased usually at the expense of the melt flow characteristics and impact properties. In the present investigations the mechanism of the stiffness and strength reinforcement has been studied and a model is proposed. This has been checked against experimental measurements and some general observations on optimization of these materials are discussed.

2. Theory of reinforcement

The most straightforward case is that of an elastic matrix reinforced by an elastic fibre which is continuous and uniaxially aligned. Then the mechanical properties in the direction of the fibres are given by a simple law of mixtures [1] as shown in Equations 1 and 2.

$$E_c = E_f V_f + E_m V_m \quad (1)$$

$$\sigma_{uc} = \sigma_{uf} V_f + \sigma_m' V_m \quad (2)$$

$$\sigma_m' = E_m \sigma_{uc} \quad (3)$$

Equations 1 and 2 may be rewritten in the form of Equations 1a and 2a

$$E_c = E_f V_f + E_m (1 - V_f) \quad (1a)$$

$$\sigma_{uc} = \sigma_{uf} V_f + E_m \epsilon_{uc} (1 - V_f) \quad (2a)$$

In the case of misaligned fibres or where the direction of testing is not coincident with the fibre axis, then the fibre contribution in Equations

1a and 2a must be reduced. For fibres randomly disposed in a plane the term is reduced to a third, and if the fibres are randomly disposed in three dimensions the factor is one sixth [2]. We may thus use a modified form of Equations 1a and 2a which contain orientation factor K whose value may lie between 1 and 0.167.

$$E_c = K E_f V_f + E_m(1 - V_f) \quad (4)$$

$$\sigma_{uc} = K \sigma_{uf} V_f + E_m \epsilon_{uc}(1 - V_f) \quad (5)$$

If the fibres are discontinuous then the fibre may carry stress only by a shear transfer process at the interface. Kelly and Tyson [3] have proposed a model where there is a linear transfer of stress from the tip of the fibre to a maximum value when the strain in the fibre is equal to that in the matrix. This has been shown to be a good approximation for metallic [1, 3] and polymer [1, 4, 5] matrices. Equation 6 relates the maximum stress in the fibre to the fibre radius and the shear strength of the fibre matrix interface.

$$\frac{L}{\sigma_f} = \frac{r}{\tau} \quad (6)$$

This leads to the concept of a critical fibre length, L_c , being that length which is necessary for the maximum stress in the fibre to reach a fibre fracture stress σ_{uf} . This may be written in the form of Equation 7.

$$L_c = \frac{\sigma_{uf} r}{\tau} = \frac{E_f \epsilon_{uc} r}{\tau} \quad (7)$$

It should be noted that if the fibre length is less than the critical length then the average stress in the fibre at composite failure is given by Equation 8 and this is half the maximum stress in the fibre.

$$\bar{\sigma}_f = \frac{L\tau}{2r} \quad (8)$$

If the fibre length is greater than the critical length, then the average stress in the fibre is given by Equation 9

$$\bar{\sigma}_f' = E_f \epsilon_c \left(1 - \frac{E_f \epsilon_c r}{2L\tau} \right) \quad (9)$$

The basis of the succeeding argument is that at any value of composite strain there is a critical fibre length L_c and fibres shorter than this will carry an average stress as given in Equation 8 which will always be less than $\frac{1}{2}E_f \epsilon_c$. Fibres longer than this carry an average stress as indicated in Equation 9 which will always be

greater than $\frac{1}{2}E_f \epsilon_c$, L_c will be given by Equation 10.

$$L_c = \frac{E_f \epsilon_c r}{\tau} \quad (10)$$

If the fibres are misoriented then a correction factor of similar form to that given in Equations 4 and 5 must be used.

Composites of the type considered contain a spectrum of fibres of different lengths. At low strains all fibres will make a contribution to the reinforcement as given by Equation 9 since L_c will be small, as the strain is increased, a progressively smaller proportion of the fibres will reinforce according to Equation 9 and an increasing proportion will follow Equation 8. We may therefore expect the slope of the load extension curve for such a material to decrease as the extension (strain) is increased. A mathematical model of this behaviour may be constructed by combining the concepts of Equations 5, 8, 9 and 10.

This equation is shown schematically as Equation 11

$$\sigma_c = CX + CY + Z \quad (11)$$

The first term is the contribution of the subcritical fibres, the second term that of the supercritical fibres, and the third term that of the matrix. The individual terms are expanded in Equations 12, 13 and 14.

$$X = \sum_{L_i < L_c} \frac{\tau L_i V_i}{2r} \quad (12)$$

$$Y = \sum_{L_j > L_c} E_f \epsilon_c \left(1 - \frac{E_f \epsilon_c r}{2L_j \tau} \right) V_j \quad (13)$$

$$Z = E_m \epsilon_c (1 - V_f) \quad (14)$$

The spectrum of fibre lengths in the composite is considered to be divided into subfractions. Subcritical subfractions are denoted L_i and their respective volume fractions V_i whilst supercritical fractions are denoted L_j and V_j . These three terms may then be combined as shown in Equation 15.

$$\sigma_c = C \left[\sum \frac{\tau L_i V_i}{2r} + \sum E_f \epsilon_c \left(1 - \frac{E_f \epsilon_c r}{2L_j \tau} \right) V_j \right] + E_m \epsilon_c (1 - V_f) \quad (15)$$

In practical systems the terms E_f , E_m and r can be readily obtained and the relationships between σ_c and ϵ_c may be obtained from a tensile test. C

and τ are however generally not known, but the fibre length distribution can be determined. If we make the assumption that the orientation factor C is independent of strain and is the same for all fibre lengths at least at small strains, then this model allows both C and τ to be determined from tensile test data.

3. Experimental

The materials used were various blends of short glass fibres in nylon 6.6 and polypropylene matrices. The nylon based composites consisted of two commercial materials from which blends were made with pure nylon so as to obtain a series of materials of varying V_f . Other glass nylon blends were also prepared in the Laboratory. Two types of polypropylene were used, one was a standard commercial material which was understood to have a low adhesion to glass whilst the other was a specially modified copolymer with good adhesion to glass [6]. Details of these materials are shown in Table I. Tensile test pieces were prepared by injection moulding using an end gate, a process which gives a good degree of alignment of the glass as

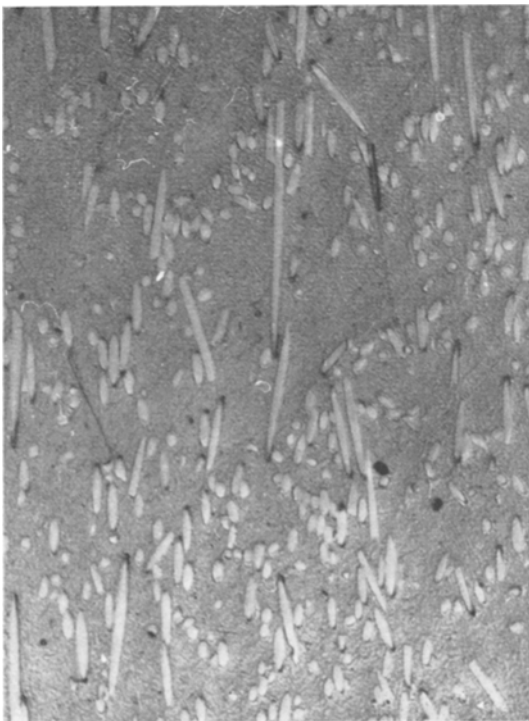


Figure 1 Longitudinal section through a glass/nylon specimen.

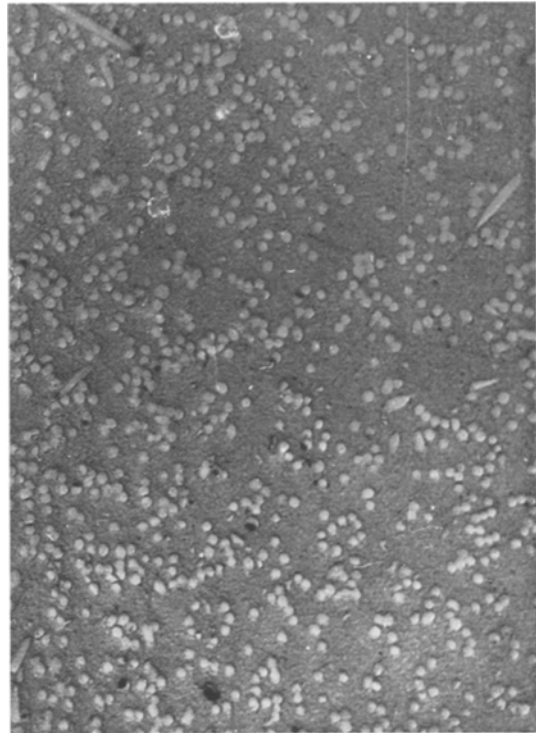


Figure 2 Transverse section through a glass/nylon specimen.

shown in Figs. 1 and 2, and also considerable molecular alignment of the polymer matrix along the axis of the test piece.

All test pieces were stored in an air conditioned laboratory atmosphere for one month before being subjected to tensile testing. This was carried out on an Instron testing machine using a sensitive strain gauge type extensometer. The fibre length distribution in each batch of material was determined by burning off the polymer, examining the fibres under the microscope and counting the fibres of various lengths in the samples. Approximately 1000 fibres were counted for each determination.

A computer programme was used to determine C and τ as follows:

- (i) A value of 69 GN/m^2 was assumed for E_f [8].
- (ii) Values of ϵ_1 and ϵ_2 were selected so that $\epsilon_2 = 2\epsilon_1$.
- (iii) The stresses σ_1 and σ_2 corresponding to the strains ϵ_1 and ϵ_2 were determined from the tensile test data. The matrix contribution, Z , (Equation 14) was calculated and the ratio R determined.

$$R = \frac{\sigma_1 - Z_1}{\sigma_2 - Z_2}$$

TABLE I Details of materials evaluated

| Composite | Matrix | Fibre | Comments |
|---------------------------|---|--|---|
| Glass/nylon A | Nylon 6.6. ICI Maranyl A100 | E glass 12 μm diam. origin unknown | Blends of ICI Maranyl A190 (0.18 V_f Glass in Maranyl A100). Maranyl AD390 (0.23 V_f Glass in Maranyl A100) and Maranyl A100 |
| Glass/nylon B | Nylon 6.6 ICI Maranyl A100 | E glass 12 μm diam. size PVA/methacrylo- chromic chloride | Blends made in our laboratory using a method similar to that used at ICI [7] |
| Glass/ polypropylene | Polypropylene copolymer ICI 8639 | E glass 12 μm diam. Fibreglass Ltd Size ref. MSS 1607 | Blends made in our laboratory by a process which will be described elsewhere |
| Glass/ polypropylene D | Polypropylene homopolymer ICI 543 | E glass 12 μm diam. Fibreglass Ltd Size ref. MSS 1607 | Blends made in our laboratory by a process which will be described elsewhere |

This is the ratio of the fibre load bearing contributions at the two selected strains. (iv) An assumed value of τ was taken and the corresponding values of L_{e_1} and L_{e_2} calculated according to Equation 10. (v) The fibre contribution terms X and Y in Equations 12 and 13 were then evaluated using the assumed values of τ and corresponding L_{e_1} and L_{e_2} for the measured fibre length distributions. (vi) These assumed values were then used to calculate R'

$$R' = \frac{X_1 + Y_1}{X_2 + Y_2}$$

(vii) The assumed value of τ was then adjusted until $R' = R$. (viii) This value of τ was then assumed to be correct and C was determined by using this value in Equation 15.

This method has proved to be very consistent.

4. Results and discussion

A typical tensile stress/strain curve for a glass filled nylon specimen is shown in Fig. 3. The majority of these specimens failed in the strain range 0.02 to 0.03 which has allowed strains of 0.01 and 0.02 to be used in the computation.

The glass filled polypropylene specimens showed a marked dependence of strain to fracture on volume fraction of filler. All specimens containing 0.15 volume fraction failed at strains of less than 0.01. For this reason only results from specimens containing less than 0.15 volume fraction glass have been used and strains of 0.005 and 0.01 have been selected for ϵ_1 and ϵ_2 respectively.

Stresses at the two levels of strain are plotted

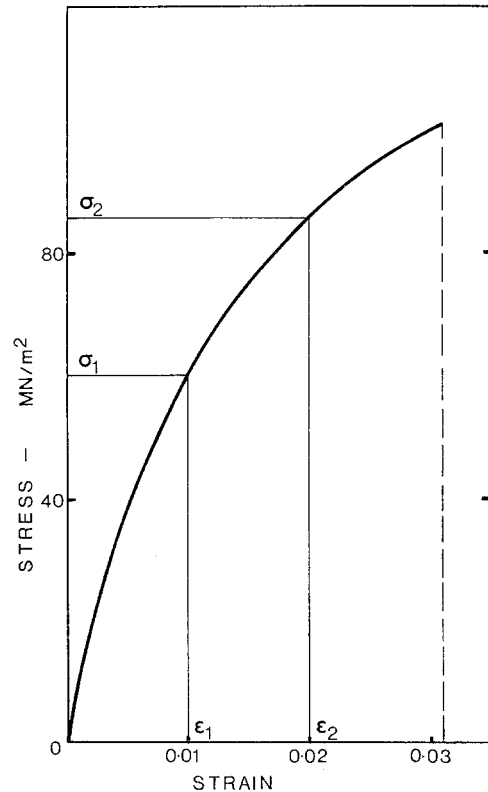


Figure 3 A typical stress/strain curve for a glass/nylon specimen.

versus volume fraction glass for the four materials in Figs. 4, 5, 6 and 7.

It can be seen that linear relationships are followed between stiffness and volume fraction (in the composition range examined); this is

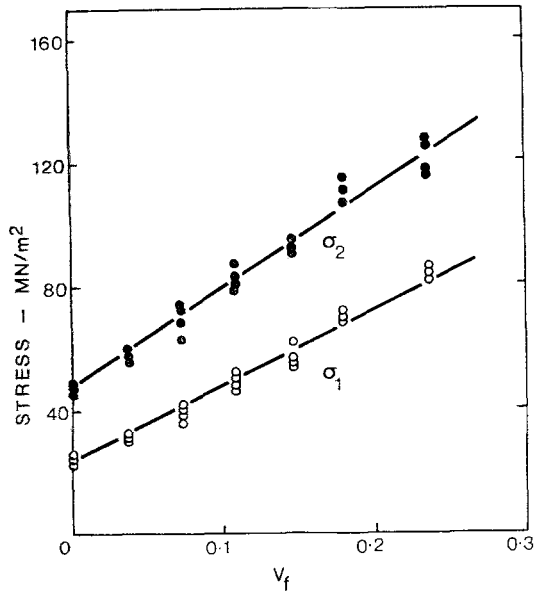


Figure 4 Stress at strains of 0.01 and 0.02 versus V_f for glass/nylon, A material.

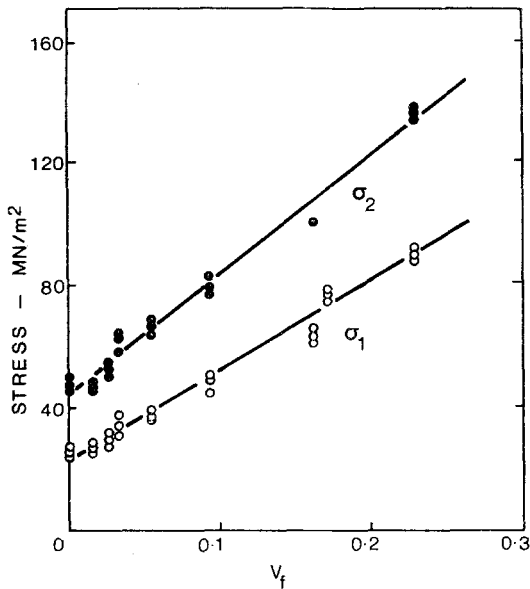


Figure 5 Stress at strains of 0.01 and 0.02 versus V_f for glass/nylon, B material.

what would be expected from Equation 15. Table II shows the calculated values of τ and C together with reinforcement efficiency factors calculated from the data on Figs. 4 to 7.

These factors have been calculated by using Equations 4 and 5. The value of C has been used to estimate the maximum advantage which might

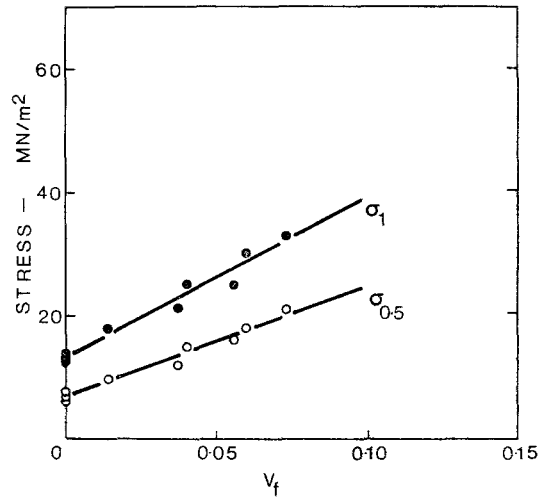


Figure 6 Stress at strains of 0.005 and 0.01 for glass polypropylene, C material.

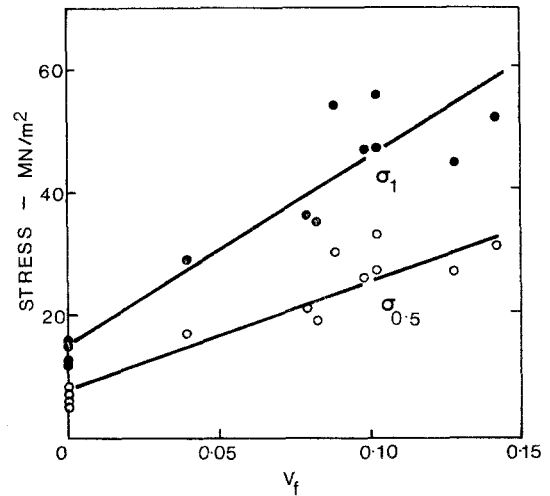


Figure 7 Stress at strains of 0.005 and 0.01 for glass polypropylene, D material.

be gained from fibres oriented in this way. Theoretical results have then been compared with the ones encountered in practice. The strain to fracture of E glass has been taken as 0.03 and the quoted strengthening efficiencies are the arithmetic mean of all specimens in the group.

The efficiency of the fibres as stiffeners decreases with increasing strain as is expected from the proposed model. The actual level of their efficiency both as stiffeners and as strengtheners, however, is poor.

In any attempt to improve the performance of short fibre reinforced composites the variables at

TABLE II

| Material | τ MN/m ² | C | Efficiency of fibre property utilization % | | |
|----------------------------------|-----------------------------|------|--|--------------|----------|
| | | | ϵ_1 | ϵ_2 | Fracture |
| Commercial glass/nylon | 45 | 0.57 | 68 | 48 | 27 |
| Laboratory glass/nylon | 50 | 0.66 | 69 | 48 | 31 |
| Glass uncoupled polypropylene | 16 | 0.75 | 77 | 50 | 10 |
| Glass coupled polypropylene | 25 | 0.68 | 81 | 67 | 30 |

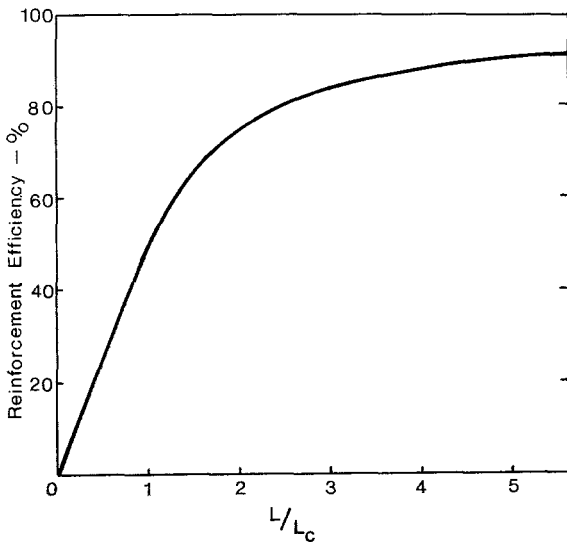


Figure 8 Proportion by volume of fibres of length greater than L versus L for the four materials.

ones disposal are V_f , τ , and the fibre length and orientation.

It may be observed from Figs. 4 to 7 that the actual stiffness may be increased by increasing V_f . There is, however, a disadvantage in doing this as the melt flow properties of the composite deteriorate and it becomes more difficult to mould. It has also been observed that the strain at fracture tends to be reduced at high V_f especially in the case of the polypropylene composites.

Efficiency could be further improved by increasing τ and this would also lower L_c . However, the values of τ determined in the glass/nylon composites are some 80% of the tensile strength of the unfilled polymer, and slightly less in the case of the coupled polypropylene.

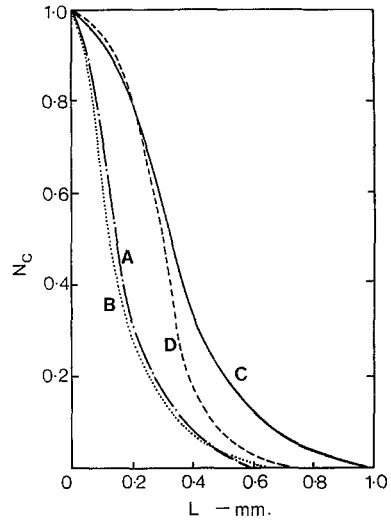


Figure 9 Fibre reinforcement efficiency versus L/L_c .

Further substantial improvement in τ is hardly feasible.

This leaves the fibre length as the only practicable variable.

The values of L_e in Table III have been

TABLE III

| Material | L_e (mm) | | |
|-----------------------|-------------------|-------------------|-------------------|
| | $\epsilon = 0.01$ | $\epsilon = 0.02$ | $\epsilon = 0.03$ |
| Glass/nylon A | 0.09 | 0.19 | 0.27 |
| Glass/nylon B | 0.08 | 0.16 | 0.25 |
| Glass/polypropylene C | 0.17 | 0.33 | 0.5 |
| Glass/polypropylene D | 0.26 | 0.52 | 0.78 |

calculated by substituting the values of τ as determined in Equation 10.

It should be noted that fibres of length L_e are loaded to the peak stress only at their centres and that the average stress in such fibres is only half the peak stress. Such fibres may be regarded as being 50% efficient and it is a consequence of Equation 9 that as fibre length increases above L_e , efficiency increases as shown in Fig. 8. Increases in length from L_e to $5L_e$ pay useful dividends in improved efficiency but further

increases in length above $5L_c$ are relatively unimportant.

The fibre distributions measured in the composites are shown in Fig. 9. The quantity N_c is the proportion of fibres by volume of length greater than L .

If we take the fracture strain of E glass to be 0.03 the values of L_c for $\epsilon = 0.03$ in Table III may be taken as L_c values. Comparing these with the fibre length distributions in Fig. 9 it can be seen that all fibres in material D and 80% of fibres in materials A, B and C are shorter than L_c . The remaining 20% of the fibres in materials A, B and C have lengths lying in the range L_c to $3L_c$.

We may expect therefore that the majority of fibres in all four materials will pull out of the matrix rather than fracture when the specimen fails. If the composites strain to the fibre fracture strain before failure then the super-critical fibres will fracture but their efficiency will always be less than 83% (corresponding to $L = 3L_c$). On the other hand if a matrix fails before the fracture strain of the few super-critical fibres is reached then the potential efficiency of the longer fibres would never be realized.

The fracture strains and strengthening efficiencies observed are consistent with failure caused by, fibre fracture in the case of the nylon based materials and matrix failure in the case of the polypropylene based materials.

It should be noted that the fibres in the mouldings tested were comparatively well aligned in the testing direction as shown in Figs. 1 and 2 and by the values of C in Table II. Due regard should be paid to this in designing mouldings since the factor C might be expected to fall to 0.167 if the fibres were randomly disposed.

5. Conclusions

1. The model treatment proposed in this work has given reliable values to fibre matrix bond strengths in the materials examined.
2. Improvements in the fibre matrix bond strength alone has led to small improvements in

stiffness and strength in glass filled nylon, and small improvements in stiffness in glass filled polypropylene.

3. The effect of fibres which are too short to be strained coherently with the matrix in polypropylene has been to stiffen but not to strengthen. This has led to failure at very low strains preventing the potential of the longer fibres from being realized.

4. Further improvements in fibre matrix bond strengths may lead to slight improvements in performance.

5. Major improvements in performance of both systems would only be achieved by increasing the fibre length to 1 to 1.5 mm for nylon and 2.5 to 4 mm for polypropylene. The very short fibres should also be eliminated if full strengthening potential is to be realized.

6. There is no point in using stronger fibres to reinforce nylon or polypropylene unless they can be compounded into the matrix without causing excessive fibre breakdown.

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