JOURNAL OF MATERIALS SCIENCE 29 (1994) 2795–2800

A modified rule-of-mixtures for prediction of tensile strengths of unidirectional fibre-reinforced composite materials

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Measured ultimate tensile strengths in unidirectional fibre-reinforced composite materials have been observed to deviate from the linear predictions of the classical rule-of-mixtures relationship. The physical factors responsible are fibre–fibre interaction, inhomogeneous fibre distribution in the matrix and fibre misorientation to the loading direction. A recent modification to the classic rule-of-mixtures to account for fibre–fibre interaction has already resulted in good agreement between measured and predicted values of ultimate tensile strengths at high fibre volume fractions for Kevlar/epoxy composites. Additional modifications to the rule of mixtures to incorporate both fibre misorientation and inhomogeneous spread have been presented here. These modifications result in greater agreement between measured and predicted ultimate tensile strengths at low fibre volume fractions while retaining the accuracy of prediction at higher fibre volume fractions. Good agreement between measured and predicted values of inhomogeneous fibre spread were obtained at high fibre volume fractions. Furthermore, these additions to the classic rule-of-mixtures can be used to gauge the extent of each of the physical factors responsible for ultimate tensile strength reduction in unidirectional composite materials.

1. Introduction

Fibre-reinforced composites are being increasingly used owing to their potential for weight reduction, enhanced strength and stiffness, and improved reliability. Fibre-reinforced laminates enable tailoring of strength and stiffness properties to meet specific structural needs. As the simplest unit of these laminates, individual laminae have been widely studied theoretically, and experimentally characterized in order to understand their behaviour under the action of applied external forces. Excellent mechanical properties are obtained when the laminae are loaded in a direction parallel to that of the fibres reinforcing them.

Knowledge of the relative proportions and the material properties and the respective properties of the two constituents in the composite material, namely the fibres and the matrix, is sufficient to predict the mechanical behaviour of unidirectional composite laminae subjected to simple tensile or compressive loading. One of the oldest and most widely used models for predicting the ultimate strength of the composite in tensile loading has been the simple rule-of-mixtures which is given in Equation 1 [1–3]

$$\sigma_c = \sigma_m V_m + \sigma_f V_f \quad (1)$$

where V_m and V_f represent the volume fractions of the matrix and the fibres, respectively, in the composite, and σ_m and σ_f are their respective strengths, with σ_c being the strength of the composite material.

When the failure strain of the fibres in the unidirectional composite is attained and the fibre volume fraction is greater than a certain minimum value (needed for the reinforcing effect of the fibres to have a positive influence on the composite strength), the ultimate composite strength is given by

$$\sigma_{cu} = \sigma_m' V_m + \sigma_{fu} V_f \quad (2)$$

where σ_{cu}, σ_m' and σ_{fu} represent the ultimate composite strength, the stress in the matrix at the failure strain of the fibres, and the ultimate fibre strength, respectively.

The above rule has some important underlying assumptions. They are [4]:

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(a) the fibres are uniformly distributed within the matrix, i.e. there are no fibre-rich or matrix-rich regions in the composite;

(b) there is perfect bonding between the matrix and the fibres;

(c) the composite is free of voids;

(d) both the fibres and the matrix behave as perfectly linear elastic materials.

Some of the practical factors influencing the longitudinal strengths of unidirectional composites are misorientation of fibres, fibre strength distribution, discontinuous fibres, stress concentrations, differential matrix reinforcement by the fibres, interface conditions and residual stresses [1]. Combinations of one or all of these factors result in the measured strengths deviating from those predicted using Equation 2. Most of these factors can be quantitatively determined. For instance, by using photomicrographs of the composite cross-section, it is possible to determine the void content, the fibre volume fraction, the fibre misorientation and inhomogeneous spread, and the interaction between fibres. Stereological and other techniques for estimating each of the above quantities have long since been established [5]. Measured longitudinal tensile strengths have been observed to deviate from the rule-of-mixtures. The deviation is a distinctly non-linear one with a negative departure from the linear values predicted by Equation 2 with the increase in fibre volume fraction, V_f. This has been observed in a wide variety of fibre/matrix composite systems [6–10]. Recently, Karam [11] has proposed a modification to the rule-of-mixtures in Equation 2 to account for fibre–fibre interaction. While the details can be found in later sections, it suffices here to say that the modified rule accurately predicts the longitudinal ultimate tensile strength variation with fibre volume fraction in carbon fibre/epoxy matrix composites [4]. Rangaraj [12] has used this modified rule to predict tensile strengths in carbon fibre/magnesium matrix composites.

The predictions made by this modified rule become less accurate in predicting strengths as the fibre volume fraction reduces. The reasons are because fibre misorientation and inhomogeneous fibre distribution have not been accounted for in this model, whereas they become important strength-reducing mechanisms in the lower fibre volume ranges.

It was the objective of the present work to improve upon Karam's model by incorporating both of the aforementioned factors and to use the result to predict longitudinal tensile strengths of a unidirectional composite material. Comparisons are then drawn between the present model and Karam's modification to the simple rule of mixtures to determine their suitability in predicting ultimate tensile strengths of unidirectional composite materials.

2. Karam's modifications to the rule of mixtures: a critique

In real unidirectional composites at high fibre volume fractions, fibre–fibre interaction in the form of overlapping and physical contact between adjacent fibres occurs. This leads to reduced interfacial surface areas between the fibres and the matrix which, in turn, results in the deviation of tensile strength from those predicted by the rule-of-mixtures given in Equation 2.

By assuming a hexagonal shape for fibre cross-sections in an idealized situation and using probability theory, Karam arrived at the following expression

$$V_{f,\,eff} = V_f(1 - V_f^2) \quad (3)$$

where $V_{f,\,eff}$ and V_f represent the effective and the actual fibre volume fractions in the composite.

In real unidirectional composites, the situation is different from that given above in that the homogeneity of fibre spread in the matrix is controlled by the manufacturing process, fibres separated by distances which are fractions of the fibre diameter create a void between them due to lack of penetration by the matrix material, and a fraction of the fibres is aligned in directions at an angle to the desired direction. This misorientation causes individual fibres to be subjected to local stress states which are different from the global stress state. This results in their premature failure and non-participation in the load-bearing process. Fibres are extremely sensitive to misorientations and even small deviations from the desired direction can lead to their failure by shear when unidirectional composites are loaded in tension [7].

In order to account for fibre–fibre interactions, a quantity termed as the "fibre interaction probability ratio", n [11], is used to relate the amount of non-load-bearing fibres to the quantity V_f^2, i.e. the relation between fibre volume fraction, V_f, and n is

$$V_{f,\,eff} = V_f(1 - n^2V_f^2) \quad (4)$$

Upon substitution of Equation 4 into the fibre strength contribution part of Equation 2, the modification to the rule-of-mixtures is obtained. The modified equation is

$$\sigma_{cu} = \sigma_m' V_m + \sigma_{fu}V_f(1 - n^2V_f^2) \quad (5)$$

Comparison between measured and predicted values of tensile strength using Equation 2 leads to the following equation for n

$$\Delta\sigma_{cu} = \sigma_{fu}n^2V_f^3 \quad (6)$$

which gives

$$n^2 = \frac{\Delta\sigma_{cu}}{\sigma_{fu}V_f^3} \quad (7)$$

where $\Delta\sigma_{cu}$ is the difference between measured and predicted composite ultimate tensile strengths from Equation 2 and the rest of the symbols have the same meanings as before.

A simple technique of estimating n from micrographs of composite laminae cross-sections taken at right angles to the fibre orientation directions has also been provided elsewhere [11]. Using quantitative stereology techniques, the following equation has been derived to compute n.

$$n^2 = \frac{S_f}{S_t}\frac{1}{V_f^2} \quad (8)$$

where S_f and S_t are the surface of the fibres in the micrograph free from contact with the matrix and the total external surface of the fibres, respectively. Each of these two quantities can be established by drawing a line of length L on the micrograph and determining the number of times, N, the line intersects the desired surface. The value of S for that surface is then given by

$$S = 2\frac{N}{L} \tag{9}$$

Using Equation 7, differences between measured and predicted values of ultimate tensile strengths from Equation 2, values of n are calculated. Using a constant value of n is necessary in Equation 5 because it is assumed to be constant there. A value of n from a certain V_f range is then chosen and used in conjunction with pre-determined values of σ_m' and σ_{fu} in Equation 5 to estimate ultimate longitudinal tensile strengths.

3. The proposed model

3.1. Inhomogeneity of fibre spread

The model given in the last section does not account for fibre misorientation and inhomogeneity of fibre spread. A simple modification to Equation 5 is proposed in this section to account for both of these factors.

A schematic drawing of the cross-section of a real composite material at right angles to the fibre/loading direction showing differential reinforcement of the matrix by the fibres is shown in Fig. 1. As shown, certain areas of the composite cross-section which are much larger than the average fibre diameter have no reinforcement. When the composite is loaded, these areas behave as a pure matrix material. In other words, the composite behaves as a two-material system, one being the composite itself and the second being the unreinforced matrix material or the matrix material which does not feel the effect of the fibre reinforcement.

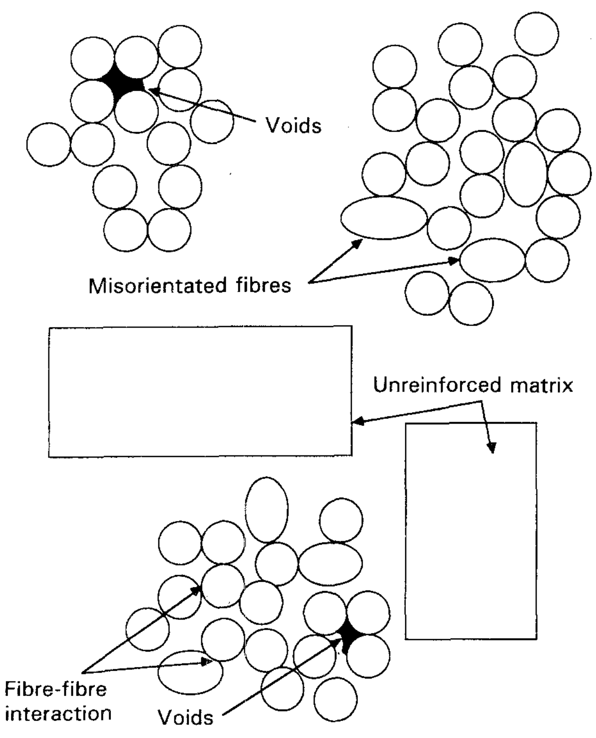

Figure 1 Schematic drawing of the cross-section of a unidirectional fibre-reinforced composite material.

Denoting the total area of the unreinforced matrix as A_{un}, we have

$$A_{eff} = A_{total} - A_{un} \tag{10}$$

where A_{eff} and A_{total} represent the area which has fibre reinforcement and the total cross-sectional area of the composite, respectively. If the two-material system thus described is subjected to the same strain, then the strength of the system is the sum of the strengths of the two materials.

If the ratio of A_{eff} to A_{total} is x, then the ratio of A_{un} to A_{total} is $(1 - x)$ because the sum of A_{eff} and A_{un} is equal to A_{total}. Further, the new fibre volume fraction in the area A_{eff} is V_f' which is related to the nominal fibre volume fraction, V_f, of the composite by the relation

$$x = \frac{V_f}{V_f'} \tag{11}$$

and the new value of n (denoted n') is

$$n'^2 = \frac{\Delta\sigma_{cu}}{\sigma_{fu} V_f'^3} \tag{12}$$

When Equation 11 is substituted into Equation 12, the expression for n' in terms of x is

$$n'^2 = \frac{x^3 \Delta\sigma_{cu}}{\sigma_{fu} V_f^3} \tag{13}$$

The relation between n and n' can be obtained by dividing Equation 13 by Equation 7. We obtain

$$n'^2 = x^3 n^2 \tag{14}$$

Using Equations 11, 13 and 14 in Equation 5 and simplifying, we obtain the modified rule for the ultimate tensile strength of unidirectional composites as

$$\sigma_{cu} = x[\sigma_m'(1 - V_f/x) + \sigma_{fu}(V_f/x)(1 - n^2 x V_f^2)] + (1 - x)\sigma_m^* \tag{15}$$

where the second part of the equation is the strength of the matrix material in the composite which does not feel the fibre reinforcement, with σ_m^* being the strength of the matrix material tested individually. Solving for x in terms of V_f, we get

$$x = \frac{V_f(\sigma_{fu} - \sigma_m') + \sigma_m^* - \sigma_{cu}}{n^2 V_f^3 \sigma_{fu} - (\sigma_m' - \sigma_m^*)} \tag{16}$$

We call x the "fibre distribution skewness factor". When using this model, the values of n are computed using either technique described in the previous section and established for a certain fibre volume fraction range of interest. Then, using Equation 16, the fibre distribution skewness factor, x, is calculated for various values of V_f. An example is illustrated in later sections. After a value of x is fixed for an appropriate range of V_f values, the strength predictions given by Equation 15 are computed. The modification at this stage assumes that fibre–fibre interaction and inhomogeneous fibre spread in the matrix are solely responsible for deviations of measured ultimate tensile strengths from that given by the rule-of-mixtures.

3.2. Fibre misorientation

Accounting for fibre misorientation is a simple extension of the above process. If y represents the fraction of the fibres which are oriented to the loading direction so that they can participate in the load-bearing process, then the effective fibre volume fraction in Equation 15, times y, represents the actual volume fraction of the fibres after accounting for fibre misorientation and inhomogeneous fibre spread. The prediction equation for the composite ultimate tensile strength accounting for fibre misorientation is then given by

$$\sigma_{cu} = x[\sigma'_m(1 - V_f/x) + \sigma_{fu}y(V_f/x)(1 - n^2xV_f^2)] + (1 - x)\sigma_m \quad (17)$$

where y represents a fraction of the fibres which are oriented such that they are effective in carrying the applied load. The quantity y will be termed the "fibre alignment fraction" hereon in this article. Solving the above equation for y in terms of V_f leads to the following relation for y

$$y = \frac{\sigma_{cu} - \sigma'_m(1 - V_f) + x(\sigma_m^* - \sigma'_m)}{\sigma_{fu}V_f(1 - n^2xV_f^2)} \quad (18)$$

4. Verifications of the present rule and discussion of results

Mittelman and Roman [6] measured ultimate longitudinal tensile strengths of unidirectional Kevlar/epoxy composites as a function of the fibre volume fraction, V_f. Their data have also been used by Karam [11] to confirm the efficacy his modification has added to the rule-of-mixtures given in Equation 2. The same data were used to determine values of the fibre distribution skewness factor, x, using Equation 16. In performing these computations, both of the quantities σ_m^* and σ'_m were taken to be of the same magnitude. Also, changes in strength in epoxy resins, which have been reported to be a function of the volume of the material stressed [13], i.e. a size effect, have been ignored. Values of x computed as a function of fibre volume fraction, V_f, have been plotted in Fig. 2. It can be seen that values of x begin to approach a stable high fibre volume fraction range between 0.9 and 1.0. A median value of x, equal to 0.97, is shown by a straight line on this plot. Using this value of x in Equation 17, values of the composite ultimate strength were determined. These results are shown in Fig. 3 as a function of the fibre volume fraction, V_f. Also shown are the measured ultimate strengths and the predictions by the Karam's rule and also the rule-of-mixtures. As can be seen, the present model outperforms both the rule-of-mixtures and Karam's modified version at the lower fibre volume fraction ranges but at the same time performs just as well as Karam's rule in the high fibre volume fraction range ($V_f > 0.6$) where Karam's rule predicts tensile strengths very close to the measured values. Also, values of x calculated

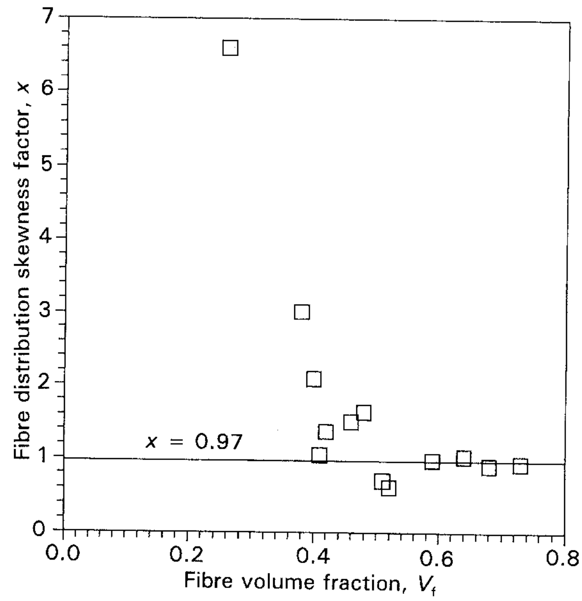

Figure 2 Variation of the fibre distribution skewness factor, x, with the fibre volume fraction, V_f.

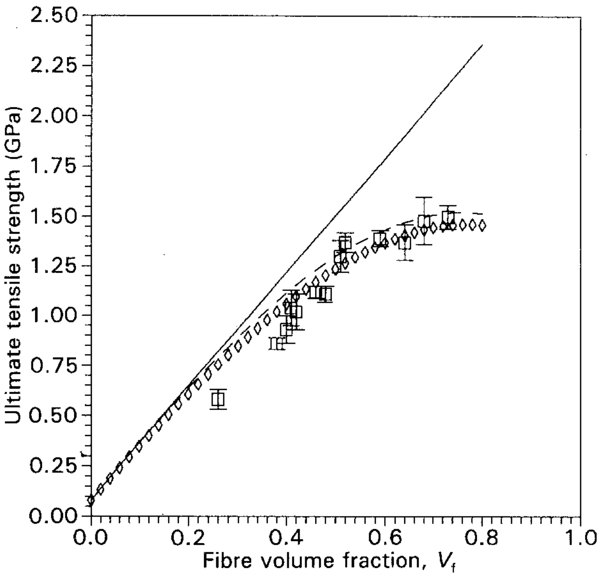

Figure 3 Comparison of (□) measured and predicted ultimate tensile strengths using (——) rule-of-mixtures, (- - -) Karam's rule and (◇) Equation 15.

indicate that at high values of V_f, the composite comes closer to becoming a one-material system, i.e. $(1 - x)$ values become very small.

Mittelman and Roman [6] also presented data showing the dependence of the coefficient of variation (CV) of fibre numbers as a function of V_f in their article. The CV was calculated using a sample population of six randomly chosen cross-sections of the composite. About 15% of the cross-sectional area of each of the samples was covered and the number of fibres enclosed in each area counted. This quantity was then non-dimensionalized by the number of fibres calculated to be present using the nominal fibre volume fraction. Their study indicated a decrease of CV with increase of V_f until a value of V_f of about 0.46, after which the CV levelled off at about 4%. It is seen that the fibre distribution skewness factor, x, at high values of V_f ($0.5 < V_f < 0.6$) is related to the CV by the simple relation

$$x = 1 - CV \quad (19)$$

when the CV is expressed as a fraction. Comparison of the values of x computed from Equation 19 show good agreement with those values shown in Fig. 2 at high values of V_f. Therefore, the simple technique used by Mittelman and Roman can be used as an experimental technique to determine x, which can then be used in Equation 15 in the absence of experimental ultimate tensile strength data.

As a next step, the fibre orientation fraction, y, was computed as a function of the fibre volume fraction, V_f, given by Equation 18 and the values of y thus obtained are shown plotted as a function of V_f, in Fig. 4. As shown, values of y for the present system increase with V_f and attain steady values for high fibre volume fractions ($V_f > 0.6$). Thus, values of n, x and y are all seen to approach unity as a high fibre volume fraction is approached. Also, at these values of V_f, the value of y indicates that for the present composite system, almost all the fibres were aligned to the loading direction within an angle (as predicted by the Halpin–Tsai equation [3]) so as to be able to participate in carrying the load, i.e. fibre misorientation becomes less of a problem in the high V_f range.

The ultimate tensile strengths calculated using Equation 18 are compared with those predicted by Karam's modification, the classical rule-of-mixtures and experimentally measured values, in Fig. 5. The use of the fibre orientation factor, y, results in predicted ultimate tensile strengths becoming closer to those measured in the lower fibre volume fraction ranges ($0.3 < V_f < 0.5$). However, at higher values of V_f, the use of y reduces the predicted tensile strengths slightly. This is due to the fact that the value of y used was more adept at predicting strengths in the lower V_f ranges. Also, as can be seen from Fig. 4, values of y in the higher V_f ranges are very close to unity and

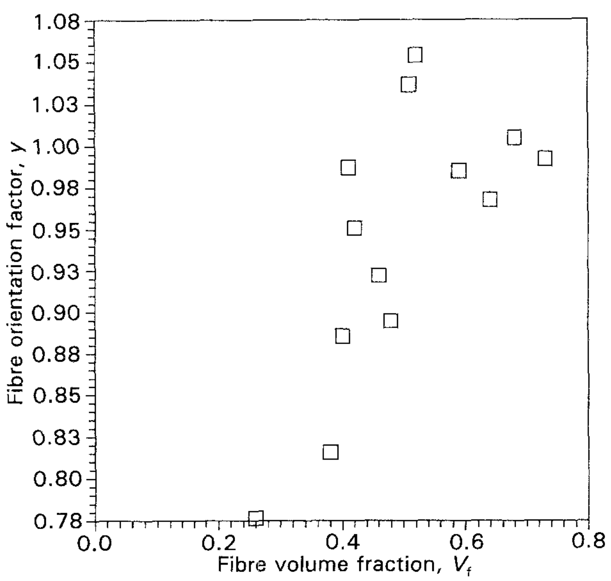

Figure 4 Variation of the fibre misorientation factor y with the fibre volume fraction V_f.

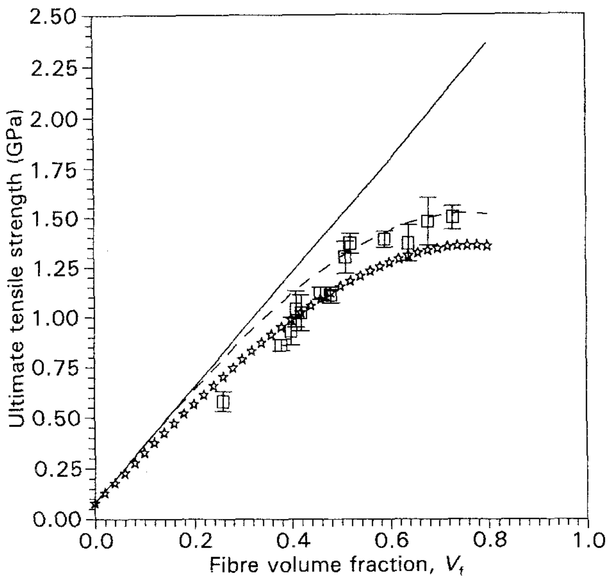

Figure 5 Comparison of (□) measured and predicted ultimate tensile strengths using (——) rule-of-mixtures, (- - -) Karam's rule and (☆) Equation 18.

therefore indicate that Equation 15 would be more appropriate in describing the strength variation for these ranges of fibre volume fractions.

Standardized stereological techniques for determination of fibre orientation angles are available. Fibre orientation distribution in continuously reinforced unidirectional composites has been recently studied by Yurgartis [14]. It has been clearly established that the fibre orientation angle distribution is normal and has been demonstrated to vary from the pre-preg to the laminate stage. Clear causes for this variation are not known, but have been attributed to be related to the manufacturing process [15]. It has also been shown that the normal distribution equations for the in-plane and out-of-plane inclination angles can be combined to obtain a binormal distribution for overall fibre orientation angle distribution. These equations would be valid for a particular range of V_f values and for a particular manufacturing process. This technique could be used to give the standard deviation of the fibre inclination angle distribution which can then be used with the cut-off inclination fibre inclination angle predicted by the Halpin–Tsai equation [3], to determine approximately the fibre alignment fraction, y, for use in strength predictions.

5. Conclusions

A modification of the non-linear relation between ultimate tensile strengths of composites and the fibre volume fraction, V_f, is presented. The modification accounts for fibre misorientation and inhomogeneous spread within the matrix. Using previously published data, the present modification has been shown to be superior to the non-linear equation suggested by Karam at low fibre volume fractions. Simple equations for determination of these two factors have also been presented. It is hoped that the use of the present equation(s) along with the accompanying stereological techniques already established can lead to reasonably close predictions of ultimate tensile strength of the composite with a prior knowledge of the quantities σ_m' and σ_{fu} without actual experimentation.

Acknowledgements

We thank C. Suryanarayana for his critical reading of the manuscript. S.B.B. acknowledges the support of NSF grant RII 8902065.

References

1. B. D. AGARWAL and L. J. BROUTMAN, "Analysis and Performance of Fiber Composites" (Wiley, New York, 1980).
2. R. M. JONES, "Mechanics of Composite Materials" (Scripta Book Company, 1975).
3. A. KELLY and N. H. MACMILLAN, "Strong Solids", 3rd Edn, (Clarendon Press, Oxford, 1986) p. 240.
4. P. K. MALLICK, in "Fiber-Reinforced Composite Materials: Manufacturing and Design" (Marcel Dekker, New York, 1988) p. 84.
5. D 3171-76 and D 3379-75, "Annual Book of ASTM Standards: Space Simulation; Aerospace Materials; High Modulus Fibers and Composites", 15.03 (American Society for Testing and Materials, Philadelphia, PA, 1983).
6. A. MITTELMAN and I. ROMAN, *Composites* **21** (1990) 63.
7. Y. KAGAWA and E. NAKATA, *J. Mater. Sci. Lett* **11** (1992) 176.
8. K. G. KRIEDER and G. R. LEVERANT, F1 in "Advanced Fibrous Reinforced Composites", Vol. 10 (Western Periodicals, N Hollywood, CA, 1966).
9. R. G. CARLSON and D. S. TOMALIN, *ibid.*, p. 45.
10. F. R. BONNANO, *ibid.*, p. 105.
11. G. N. KARAM, *Composites* **22** (1991) 84.
12. S. S. RANGARAJ, unpublished.
13. E. M. ODOM and D. F. ADAMS, *J. Mater. Sci.* **27** (1992) 1767.
14. S. W. YURGARTIS, *Compos. Sci. Technol.* **30** (1987) 279.
15. S. R. DOSHI, J. M. DEALY and J. M. CHARRIER, *Polym. Eng. Sci.* **26** (1986) 468.

Received 7 August 1992
and accepted 31 August 1993