Bounds for the elastic constants of reinforcing fibres in polymeric composites

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An analytical method is presented which enables the elastic constants of the fibres in a unidirectional composite to be estimated from a knowledge of the elastic constants of the composite and the matrix resin. Results are presented for polyethylene fibre/epoxy resin and carbon fibre/epoxy resin composites, and it is shown that the predicted fibre constants are, in both cases, close to those obtained from other methods.

1. Introduction

The calculation of the elastic constants of a fibre-reinforced composite is a very familiar exercise which is well documented in the literature (see, for example, [1, 2]). Research by the present authors and their colleagues has addressed this subject in some detail [3, 4] with particular regard to recent attempts to obtain more accurate theoretical estimates using analytical procedures [5], simple bounding methods [6] or finite element analyses [7]. It has become apparent from these recent researches that if such new theoretical techniques are to be applied effectively, it will be necessary to have accurate values for the elastic constants of the fibres. It is evident that for many of the composite systems which are of great interest, e.g. those involving carbon fibres, well authenticated accurate values do not exist.

One method reported in the literature [8] for obtaining the elastic constants of a carbon fibre involves manufacturing a group of unidirectional composites with different fibre volume fractions. The elastic properties of the composites are then measured, and the fibre properties obtained by plotting the composite results and extrapolating to 100% fibre volume fraction. To obtain accurate values for the fibre elastic constants by this method obviously requires a significant number of samples to be produced over a large range of fibre volume fractions.

The aim of the present work was to propose a new method for obtaining the fibre elastic constants from measurements of the elastic constants of a unidirectional composite. By using our analytical techniques for describing the elastic constants of a composite, it is, in principle, possible to determine the fibre elastic constants from a single composite sample.

In this paper it will be shown that the fibre properties can be determined with some accuracy, and results will be presented for polyethylene fibre/epoxy composites which have led to a set of estimate elastic constants for the polyethylene fibres, and a single carbon fibre epoxy composite, from which an estimate of the carbon fibre properties have been obtained.

2. Theory

2.1. Unidirectional fibre composites

Following the procedure adopted in a previous publication [6], the elastic constants are calculated on the basis of both series–parallel and parallel–series models. For the calculation of the extensional elastic constants, the models are shown schematically in Fig. 1 and 2, and for the shear constants in Figs 3 and 4. In our previous publication [6], bounds for the elastic constants were determined starting with the elastic properties of the matrix and the fibres, for a given volume fraction of the fibres. It was found that in most instances the calculated bounds are very close. This suggested that it would be reasonable to attempt to invert the direction of the calculations, and estimate the fibre elastic constants on the basis that the calculated bounds are made as close as possible to the measured elastic constants of the composite by optimizing the values of the elastic constants of the fibres to achieve the closest fit.

It is appropriate to separate the bounds calculations for the extensional and shear elastic constants.

2.1.1. Extensional elastic constants

The theoretical calculations for the extensional elastic constants involve four sets of equations describing four cases which are illustrated schematically in Figs 1 and 2. These four cases are:

Case 1 Series–parallel situation for loading parallel to the fibre direction (Fig. 1).
Case 2 Series–parallel for loading perpendicular to the fibre direction (Fig. 1).

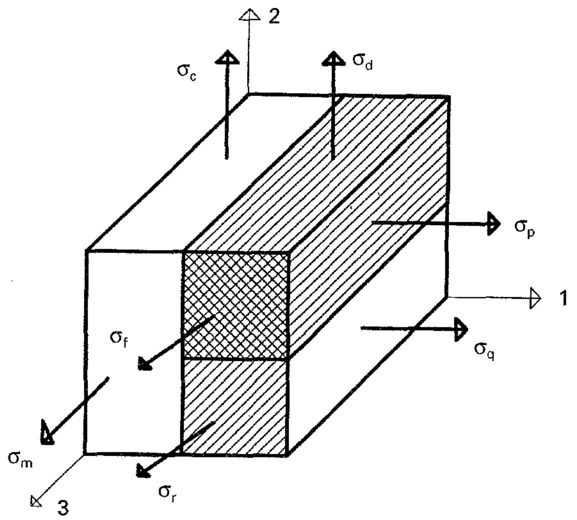

Figure 1 The series–parallel situation for extensional loading parallel and perpendicular to the fibre direction.

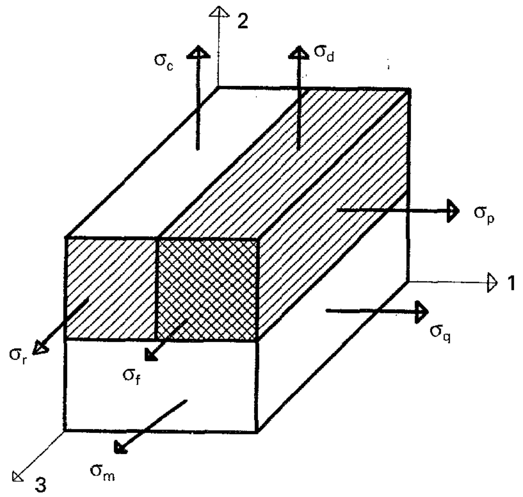

Figure 2 The parallel–series situation for extensional loading parallel and perpendicular to the fibre direction.

Case 3 Parallel–series situation for loading parallel to the fibre direction (Fig. 2).

Case 4 Parallel–series for loading perpendicular to the fibre direction (Fig. 2).

The relevant equations for these four cases will now be given.

2.1.1.1. Case 1. The equilibrium conditions for unit stress applied in the fibre direction are

$$(1 - f^{\frac{1}{2}})\sigma_m + f\sigma_f + f^{\frac{1}{2}}(1 - f^{\frac{1}{2}})\sigma_r = 1 \quad (1a)$$

$$(1 - f^{\frac{1}{2}})\sigma_c + f^{\frac{1}{2}}\sigma_d = 0 \quad (1b)$$

$$f^{\frac{1}{2}}\sigma_p + (1 - f^{\frac{1}{2}})\sigma_q = 0 \quad (1c)$$

where σ_m, σ_f and σ_r are the stresses in the fibre direction and $\sigma_c, \sigma_d, \sigma_p$ and σ_q are stresses in the transverse directions, as shown in Fig. 1: f is the fibre volume fraction.

The strains ε_{33} and ε_{13}, the strains in the fibre direction 3 and in the transverse direction 1, respec-

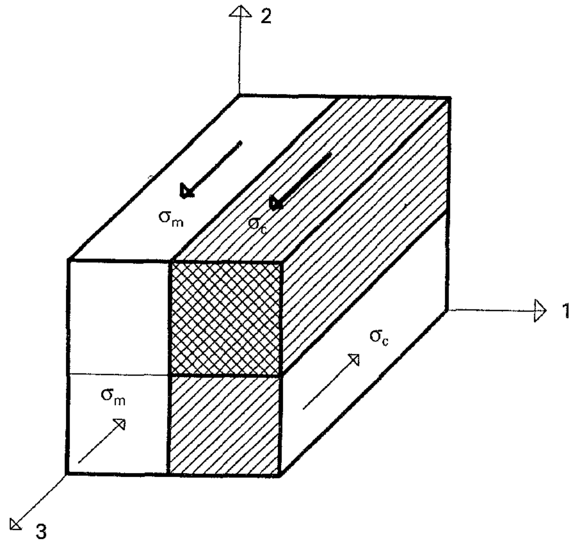

Figure 3 The series–parallel situation for shear loading parallel to the fibre direction.

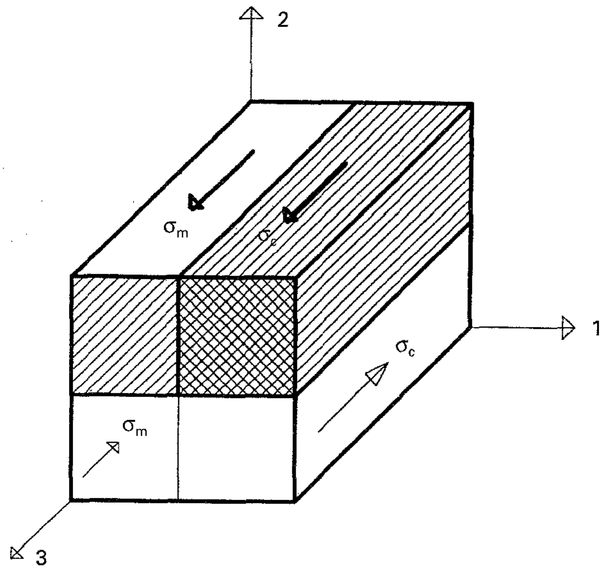

Figure 4 The parallel–series situation for shear loading perpendicular to the fibre direction.

tively, for unit stress applied in the fibre direction 3, are related to the elastic constants E_m, ν_m of the matrix by the four equations

$$\varepsilon_{33} = \frac{\sigma_m}{E_m} - \frac{\nu_m}{E_m}\sigma_c \quad (2a)$$

$$\varepsilon_{23} = \frac{\sigma_c}{E_m} - \frac{\nu_m}{E_m}\sigma_m \quad (2b)$$

$$\varepsilon_{33} = \frac{\sigma_r}{E_m} - \frac{\nu_m}{E_m}\sigma_d - \frac{\nu_m}{E_m}\sigma_q \quad (2c)$$

and

$$\varepsilon_{13} = f^{\frac{1}{2}}\left(\frac{\sigma_q}{E_m} - \frac{\nu_m}{E_m}\sigma_d - \frac{\nu_m}{E_m}\sigma_r\right) - (1 - f^{\frac{1}{2}})\left(\frac{\nu_m}{E_m}\sigma_m + \frac{\nu_m}{E_m}\sigma_c\right) \quad (2d)$$

Three further equations, based on compatibility of strains, but involving the elastic constants of the fibre can be obtained.

$$\varepsilon_{33} = s_{33}\sigma_f + s_{13}(\sigma_d + \sigma_p) \quad (3a)$$

$$s_{13}\sigma_f + s_{11}\sigma_p + s_{12}\sigma_d = \frac{1}{E_m}(\sigma_q - \nu_m\sigma_d - \nu_m\sigma_r) \quad (3b)$$

$$\varepsilon_{23} = f^{\frac{1}{2}}(s_{13}\sigma_f + s_{11}\sigma_d + s_{12}\sigma_p) + (1 - f^{\frac{1}{2}})\frac{1}{E_m}(\sigma_d - \nu_m\sigma_q - \nu_m\sigma_r) \quad (3c)$$

where the fibre elastic compliance matrix is defined by

$$\begin{pmatrix} s_{11} & s_{12} & s_{13} & 0 & 0 & 0 \\ s_{12} & s_{11} & s_{13} & 0 & 0 & 0 \\ s_{13} & s_{13} & s_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & s_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & s_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & 2(s_{11} - s_{12}) \end{pmatrix} \quad (4)$$

with the 3 direction as the fibre axis.

2.1.1.2. Case 2. The equilibrium equations for unit stress applied in the transverse direction are

$$(1 - f^{\frac{1}{2}})\sigma_m + f\sigma_f + f^{\frac{1}{2}}(1 - f^{\frac{1}{2}})\sigma_r = 0 \quad (5a)$$

$$(1 - f^{\frac{1}{2}})\sigma_c + f^{\frac{1}{2}}\sigma_d = 0 \quad (5b)$$

$$f^{\frac{1}{2}}\sigma_p + (1 - f^{\frac{1}{2}})\sigma_q = 1 \quad (5c)$$

The strains ε_{31} and ε_{21}, the strains in the fibre direction 3 and in the transverse direction 2, respectively, for unit stress applied in the transverse direction 1, are related to the elastic constants E_m, ν_m of the matrix by the four equations

$$\varepsilon_{31} = \frac{\sigma_m}{E_m} - \frac{\nu_m}{E_m} - \frac{\nu_m}{E_m}\sigma_c \quad (6a)$$

$$\varepsilon_{31} = \frac{\sigma_r}{E_m} - \frac{\nu_m}{E_m}\sigma_d - \frac{\nu_m}{E_m}\sigma_q \quad (6b)$$

$$\varepsilon_{21} = \frac{\sigma_c}{E_m} - \frac{\nu_m}{E_m} - \frac{\nu_m}{E_m}\sigma_m \quad (6c)$$

$$\varepsilon_{11} = f^{\frac{1}{2}}\left(\frac{\sigma_q}{E_m} - \frac{\nu_m}{E_m}\sigma_d - \frac{\nu_m}{E_m}\sigma_r\right) + (1 - f^{\frac{1}{2}})\left(\frac{1}{E_m} - \frac{\nu_m}{E_m}\sigma_m - \frac{\nu_m}{E_m}\sigma_c\right) \quad (6d)$$

As before, three further equations involving the elastic constants of the fibre can be obtained

$$\varepsilon_{31} = s_{33}\sigma_f + s_{13}(\sigma_d + \sigma_p) \quad (7a)$$

$$s_{13}\sigma_f + s_{11}\sigma_p + s_{12}\sigma_d = \frac{1}{E_m}(\sigma_q - \nu_m\sigma_d - \nu_m\sigma_r) \quad (7b)$$

$$\varepsilon_{21} = f^{\frac{1}{2}}(s_{13}\sigma_f + s_{11}\sigma_d + s_{12}\sigma_p) + (1 - f^{\frac{1}{2}})\frac{1}{E_m}(\sigma_d - \nu_m\sigma_q - \nu_m\sigma_r) \quad (7c)$$

2.1.1.3. Case 3. For the parallel–series situation the equilibrium equations for unit stress applied in the fibre direction are

$$(1 - f^{\frac{1}{2}})\sigma_m + f\sigma_f + f^{\frac{1}{2}}(1 - f^{\frac{1}{2}})\sigma_r = 1 \quad (8a)$$

$$(1 - f^{\frac{1}{2}})\sigma_c + f^{\frac{1}{2}}\sigma_d = 0 \quad (8b)$$

$$f^{\frac{1}{2}}\sigma_p + (1 - f^{\frac{1}{2}})\sigma_q = 0 \quad (8c)$$

where σ_m, σ_f, σ_r, etc., are the stresses shown in Fig. 1, but for this different bound will not be identical to those determined for Case 1 above.

The strains ε_{33} and ε_{13} for this bound are related to the elastic constants of the matrix by the equations

$$\varepsilon_{33} = \frac{\sigma_m}{E_m} - \frac{\nu_m}{E_m}\sigma_q \quad (9a)$$

$$\varepsilon_{13} = \frac{\sigma_q}{E_m} - \frac{\nu_m}{E_m}\sigma_m \quad (9b)$$

$$\varepsilon_{33} = \frac{\sigma_r}{E_m} - \frac{\nu_m}{E_m}\sigma_c - \frac{\nu_m}{E_m}\sigma_p \quad (9c)$$

$$\varepsilon_{23} = f^{\frac{1}{2}}\left(\frac{\sigma_c}{E_m} - \frac{\nu_m}{E_m}\sigma_r - \frac{\nu_m}{E_m}\sigma_p\right) - (1 - f^{\frac{1}{2}})\left(\frac{\nu_m}{E_m}\sigma_m + \frac{\nu_m}{E_m}\sigma_q\right) \quad (9d)$$

The three further equations, based on compatibility of strains are now

$$\varepsilon_{33} = s_{33}\sigma_f + s_{13}(\sigma_d + \sigma_p) \quad (10a)$$

$$s_{13}\sigma_f + s_{11}\sigma_d + s_{12}\sigma_p = \frac{1}{E_m}(\sigma_c - \nu_m\sigma_p - \nu_m\sigma_r) \quad (10b)$$

$$\varepsilon_{23} = f^{\frac{1}{2}}(s_{13}\sigma_f + s_{11}\sigma_p + s_{12}\sigma_d) + (1 - f^{\frac{1}{2}})\frac{1}{E_m}(\sigma_p - \nu_m\sigma_c - \nu_m\sigma_r) \quad (10c)$$

2.1.1.4. Case 4. The equilibrium equations for the parallel–series case where unit stress is applied in the transverse direction are

$$(1 - f^{\frac{1}{2}})\sigma_m + f\sigma_f + f^{\frac{1}{2}}(1 - f^{\frac{1}{2}})\sigma_r = 0 \quad (11a)$$

$$(1 - f^{\frac{1}{2}})\sigma_c + f^{\frac{1}{2}}\sigma_d = 0 \quad (11b)$$

$$f^{\frac{1}{2}}\sigma_p + (1 - f^{\frac{1}{2}})\sigma_q = 1 \quad (11c)$$

In this case, the strains ε_{11}, ε_{21} and ε_{31} are related to the elastic constants of the matrix by the equations

$$\varepsilon_{11} = \frac{\sigma_q}{E_m} - \frac{\nu_m}{E_m}\sigma_m \quad (12a)$$

$$\varepsilon_{21} = f^{\frac{1}{2}}\left(\frac{\sigma_c}{E_m} - \frac{\nu_m}{E_m}\sigma_p - \frac{\nu_m}{E_m}\sigma_r\right)$$

$$- (1 - f^{\frac{1}{2}})\left(\frac{\nu_m}{E_m}\sigma_m + \frac{\nu_m}{E_m}\sigma_q\right) \quad (12b)$$

$$\varepsilon_{31} = \frac{\sigma_m}{E_m} - \frac{\nu_m}{E_m}\sigma_q \quad (12c)$$

$$\varepsilon_{31} = \frac{\sigma_r}{E_m} - \frac{\nu_m}{E_m}\sigma_c - \frac{\nu_m}{E_m}\sigma_p \quad (12d)$$

and the three final equations, again based on compatibility of strains are

$$\varepsilon_{11} = f^{\frac{1}{2}}(s_{13}\sigma_f + s_{11}\sigma_p + s_{12}\sigma_d)$$

$$+ (1 - f^{\frac{1}{2}})\frac{1}{E_m}(\sigma_p - \nu_m\sigma_c - \nu_m\sigma_r) \quad (13a)$$

$$s_{13}\sigma_f + s_{11}\sigma_d + s_{12}\sigma_p$$

$$= \frac{1}{E_m}(\sigma_c - \nu_m\sigma_p - \nu_m\sigma_r) \quad (13b)$$

$$\varepsilon_{31} = s_{33}\sigma_f + s_{13}(\sigma_d + \sigma_p) \quad (13c)$$

2.1.2. Shear elastic constants

The series–parallel and parallel–series situations for the shear elastic constants are shown schematically in Figs 3 and 4, respectively.

For the series–parallel situation (Fig. 3) the equation of equilibrium for unit shear stress is

$$f^{\frac{1}{2}}\sigma_c + (1 - f^{\frac{1}{2}})\sigma_m = 1 \quad (14)$$

and the shear strain in the composite e_{44} is given by

$$\varepsilon_{44} = \frac{\sigma_m}{G_m}$$

$$= f^{\frac{1}{2}}s_{44}\sigma_c + (1 - f^{\frac{1}{2}})\frac{\sigma_c}{G_m} \quad (15)$$

where G_m and s_{44} are the shear modulus of the matrix and the shear compliance of the fibre, respectively.

On the assumption that the shear strain in the composite is equal to the shear strain in the matrix, it follows that for unit shear stress

$$\varepsilon_{44} = \frac{\sigma_m}{G_m} = \frac{1}{G_4} \quad (16)$$

where G_4 is the longitudinal shear modulus of the composites.

Combining Equations 14–16 gives

$$\frac{1}{s_{44}} = G_m\frac{[G_4 - G_m(1 - f^{\frac{1}{2}})]f^{\frac{1}{2}}}{G_m(1 - f^{\frac{1}{2}} + f) - G_4(1 - f^{\frac{1}{2}})} \quad (17)$$

Similarly for the parallel–series situation shown in Fig. 4, the equilibrium equation for unit shear stress is given by

$$f^{\frac{1}{2}}\sigma_c + (1 - f^{\frac{1}{2}})\sigma_m = 1 \quad (18)$$

In this case, the equation describing compatibility of strain relates only to part of the structure and gives

$$f^{\frac{1}{2}}\varepsilon_{44} = \frac{\sigma_m}{G_m} = s_{44}\sigma_c \quad (19)$$

and the composite shear modulus, G_4, is given by

$$\frac{1}{G_4} = f^{\frac{1}{2}}\frac{\sigma_m}{G_m} + (1 - f^{\frac{1}{2}})\frac{1}{G_m} \quad (20)$$

Combining Equations 18–20 gives

$$\frac{1}{s_{44}} = G_m\frac{G_4(1 - f^{\frac{1}{2}} + f) - G_m(1 - f^{\frac{1}{2}})}{[G_m - G_4(1 - f^{\frac{1}{2}})]f^{\frac{1}{2}}} \quad (21)$$

Equations 17 and 21 are the required bounds for the shear compliance of the fibre.

2.2. Numerical procedure for calculation of fibre elastic constants

In general, a set of linear algebraic equations may have an exact solution only if the number of unknowns is equal to the number of equations, otherwise only an approximate solution can be found. One of the simplest methods of reaching a solution for such a case is to minimize the errors by using the least squares method. Considering a set of equations

$$a_{ij}x_j + b_i = 0$$

where $i = 1, \ldots, m$, $j = 1, \ldots, n$, one finds in general that the right-hand side is not equal to zero. To obtain the minimum of so-defined errors one can form a sum of the squared errors,

$$(a_{ij}x_j + b_i)(a_{ik}x_k + b_i) = S$$

where $i = 1, \ldots, m$; $j, k = 1, \ldots, n$, and find the minimum of this sum by making the first differential equal to zero

$$\frac{\partial S(x_j)}{\partial x_j} = 0 \quad (22)$$

This procedure was used throughout.

Equations 1 and 2 describing Case 1 above can, in principle, be solved to eliminate the seven stresses σ_m, σ_f, etc., because the quantities ε_{33}, ε_{13} and ε_{23} define the strains for unit stress and may therefore be regarded equivalently as the corresponding elastic compliances of the composite, which are known quantities. We are therefore left with Equations 3a–c for the four unknowns s_{33}, s_{11}, s_{12} and s_{13}, the elastic compliances of the fibres. Similar considerations apply with regard to Equations 5–7 of Case 2. By taking all the six sets of Equations 1–7 together, it is possible to adopt a minimization procedure which gives one bound for the elastic constants of the fibres.

An identical procedure can be adopted to obtain the parallel–series bound for the elastic constants of the fibres, using Equations 8–13 of Cases 3 and 4.

Finally, it is possible to combine all the equations for both the parallel–series and series–parallel cases to predict final values for the elastic constants of the fibres. Where the bounds are close together (for the polyethylene fibre), only a single value is quoted: for the carbon fibre, the predicted bounds were, in some cases, further apart and the actual bounding values are therefore quoted.

3. Experimental procedure
3.1. Composites preparation
Composites were prepared following standard procedures adopted in our laboratory and described in detail elsewhere [9, 10]. The method employed was a standard prepreg route, using a drum winder and a partially cured epoxy resin, Fibredux 913 made by Ciba Geigy. For the polyethylene fibre/epoxy resin composites, the fibre was a high-modulus polyethylene fibre produced by the melt spinning/hot drawing route invented at Leeds University and commercialized by Snia Fibre (Tenfor fibres) and more recently by Hoechst–Celanese (Certran fibres). Composites were made at fibre volume fractions of nominally 40%, 50%, 55%, 60% and 65%. For the carbon fibre/epoxy composite, the fibre used was Courtaulds HM370: only one fibre volume fraction, nominally 50%, was manufactured.

3.2. Measurement of fibre elastic constants
The elastic properties of the composites were measured using an ultrasonic velocity technique. In this method, originally published by NPL [11], and developed further by Dyer *et al.* [3] and Woolf [12], the basic measurement is the time of flight for a sound pulse to travel through the sample. Measurement of the travel time at various angles of incidence allows the four elastic constants in the plane of sound propagation to be determined. By propagation in different planes, a full set of elastic constants can be determined. For the transversely isotropic composite samples under consideration here, only two experiments (propagation in the 23 and 12 planes) were needed to calculate the five independent elastic constants needed for a full description of the composite elastic behaviour. A more detailed account of this technique can be found elsewhere [3].

4. Results
4.1. Polyethylene fibre/epoxy composites
In order to assess the accuracy of the numerical technique presented in this paper for determining fibre elastic constants, it is necessary to have a fibre whose elastic properties are known. In a recent paper [13] we described how the technique of hot compaction, invented at Leeds University, can be used to determine the elastic properties of a range of thermoplastic polymer fibres. In essence, the process works by "selectively melting" a small fraction of the outer surface of a polymer fibre which, on cooling, recrystallizes to bind the structure together [14]. This process allows very high fibre volume fraction samples to be produced, close to the theoretical limit of 91%. By manufacturing a range of samples of different volume fractions, and measuring the elastic properties of the compacted materials, it is possible to extrapolate to 100% fibre properties. This procedure is very similar to that of Dean and Turner [8], described in Section 1, in their work on carbon fibre composites. The advantage of the compaction process is that very high fibre volume fractions can be achieved, giving a high degree of accuracy to the extrapolation procedure.

TABLE I A comparison of the elastic properties of high-modulus melt-spun polyethylene fibre determined by the numerical procedure and by the hot compaction route

	Numerical procedure	Hot compaction
E_{33} (GPa)	66.9 ± 6.1	68.5
E_{11} (GPa)	4.49 ± 0.31	4.71
ν_{13}	0.50 ± 0.07	0.47
ν_{12}	0.55 ± 0.02	0.58
G_{13} (GPa)	1.65 ± 0.17	1.65

TABLE II A comparison of the elastic properties of HM370 carbon fibre determined by the numerical procedure and by the best set of data combining manufacturers' data and the method of Smith [15]

	Numerical procedure			Manufacturers' data, Smith's method
	Lower bound	Upper bound	Average	
E_{33} (GPa)	381.4	381.6	381.5	370
E_{11} (GPa)	13.0	13.7	13.35	12
ν_{13}	0.41	0.41	0.41	0.35
ν_{12}	0.55	0.59	0.57	0.48
G_{13} (GPa)	11.8	23.0	17.4	17.5

Table I shows a comparison of the elastic properties of the melt-spun high-modulus polyethylene fibre, determined by the numerical technique described in this paper and the hot compaction process. The numerical technique shows results of measurements on five fibre volume fractions of polyethylene fibre/epoxy resin composites (42%, 51%, 56%, 66% and 67%). The agreement between the procedures is seen to be excellent, validating the new numerical technique.

4.2. Carbon fibre/epoxy resin composite
As a second test of the numerical method, an HM370 carbon fibre/epoxy composite was evaluated. Again the advantage of this system is that the carbon fibre elastic constants are at least partly known. We have used this fibre before in our previous work [4] evaluating different theoretical models for predicting composite properties. The carbon fibre elastic constants needed for the previous study were taken partly from the manufacturers' data sheets (longitudinal modulus, E_{33}, and Poisson's ratio, ν_{13}), and partly by using a technique reported by Smith [15]. Smith showed that plotting graphs of each fibre elastic constant against the longitudinal modulus, E_{33}, using all the available literature for carbon fibre properties, produced reasonable correlations which could then be used to predict the unknown properties of a fibre in which only the longitudinal modulus was known.

Table II shows a comparison between the elastic properties of the HM370 carbon fibre determined from the numerical technique and using the graphical method of Smith. For the numerical method, both the bounds and the average of the bounds is shown. The agreement between the two predictions is seen to be good. The bounds produced by the numerical procedure were, in general, very close together, as was seen for the polyethylene fibre, apart from the prediction of the shear modulus, G_{13}.

5. Conclusions

A new procedure for determining the elastic properties of a fibre from measurements on unidirectional composites, is described. Comparison with two known fibre systems, a polyethylene fibre/epoxy resin composite and an HM370 carbon fibre composite, suggests that the new numerical procedure is valid. It should now be possible, with some confidence, to use the procedure to determine the elastic properties of an unknown fibre.

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