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The effect of interfacial conditions on the elastic-longitudinal modulus of fibre reinforced composites

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A model study is conducted on the prediction of the elastic longitudinal modulus E_{CL} of a unidirectional fibre reinforced composite. It is assumed in our model that the fibres are aligned in the uniaxial loading direction and that the representative volume element (RVE) consists of three coaxial cylinders, namely the fibre, the interphase and the bulk matrix. The interphase represents the third phase developed between the constituent phases of the composite and it is characterized by mechanical imperfections, physicochemical interactions and limited mobility of macromolecules due to their absorption on the filler surface. Thus the interphase properties are varied within this third phase in an unknown way. In the present study it is supposed that the elastic modulus of the interphase material E_i is varied along the thickness following an exponential law of variation. Moreover, the interphase thickness is determined according to existing theory which is based on thermal capacity measurements. Predicted E_{CL} values agree well with respective experimental results. In addition the effect of an abrupt variation of the elastic modulus at the fibre surface on E_{CL} is considered. Results showed that this type of variation is of minor importance in predicting E_{CL}.

1. Introduction

In a fibre reinforced composite the fibres bear the major fraction of the load. However, one of the most important factors affecting the reinforcement mechanism is the nature and quality of the fibre–matrix adhesion [1–4]. In fact the loads are transferred from the fibres to the matrix through an interfacial region, often called the *interphase* [5–10].

In real composites, around an inclusion embedded in a matrix a rather complex situation develops, consisting of areas of imperfect bonding, mechanical stresses due to shrinkage, high stress gradients or even stress singularities due to the geometry of the inclusion, voids, microcracks etc. Thus, the presence of the reinforcement into the polymer material results in an imperfection in the polymer structure. More precisely, absorption interaction restricts the segmental mobility and affects molecular packing in the layer of the polymeric matrix which is in contact with the filler surface. In such a case the polymer between two fillers consists of (i) a boundary layer which is called *boundary interphase* or simply *interphase* with its own properties and (ii) of unchanged matrix.

Although the thickness of an interphase is of the order of several hundred nanometres, its existence is of paramount importance since it strongly affects the relaxation properties of polymeric composites. For example [11], the glass transition, T_g of filled polymers depends linearly on the fraction of the polymer, U_i, in the boundary interphase; i.e.

$$T_{gf} = T_{go} + \Delta T U_i \tag{1}$$

where T_g is the glass transition temperature, the indices f and o refer to the filled and unfilled polymer respectively and ΔT is a constant corresponding to the increase in T_g for the system where all of the polymer is in the boundary layer ($U_i = 1$). Also it was established [11] that in the case where T_g values of the polymer matrix and interphase material differ by 20 to 40° C then two T_g values corresponding to the unchanged polymer matrix and the boundary interphase may be experimentally observed. This depends on the boundary layer concentration – which in turn depends on filler concentration – and on the conditions of deformation.

Especially in the case of short fibre composites, the interphase determines the *critical length* and hence the efficiency of reinforcement. Moreover, it plays an important role on the shear and off-axis properties of aligned fibre composites [12].

Interfacial conditions strongly affect the tensile behaviour of unidirectional metal matrix composites [13–15]. In this type of composites interfacial reaction is one of the main reasons for reducing composite strength [16–22].

It is possible by means of thermal capacity measurements to determine the effective thickness of the boundary interphase. [23]. More precisely, as the filler

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volume fraction is increased, the jump in thermal capacity ΔC_p in the glass transition region is reduced. This is a definite indication that a certain number of macromolecules close to the rigid surface of the inclusions is excluded from the cooperative process of the glass transition. The portion of these macromolecules, λ, is given by the relation

$$\lambda = 1 - \frac{\Delta C_p^f}{\Delta C_p^0} \qquad (2)$$

where ΔC_p^f and ΔC_p^0 is the thermal capacity jump for the filled and unfilled polymer respectively. Then, the thickness of the boundary interphase can be found from the equation

$$\left(\frac{\Delta r + r_f}{r_f}\right)^3 - 1 = \lambda \frac{U_f}{1 - U_f} \qquad (3)$$

where r_f, Δr and U_f stand for the fibre, the interphase thickness and the fibre volume fraction respectively.

Apart from the above described experimental method, a large number of other experimental methods, usually spectroscopic, have been developed for determining the extent of the boundary interphase. Also, mathematical models have been developed to predict the mechanical and/or thermal behaviour of FRP. A brief outline of the various predictive theories is given by Paipetis [24]. In all these models, several general assumptions were made such as : (1) The ply is macroscopically homogeneous, linearly elastic and generally orthotropic or transversely isotropic; (2) the fibres are linearly elastic and homogeneous; (3) the matrix material is linearly elastic and homogeneous; (4) the fibre and matrix are free of voids; (5) there is complete bonding at the interface of the constituent materials and there is no transition region between them; (6) the ply is initially in a stress free state; and (7) the fibres are either regularly spaced and/or aligned.

Models can be divided into three main categories depending on the method followed in developing each one of them; namely: (a) *self consistent model methods* [25–35]; (b) *variational methods* [36–41]: (c) *exact methods* [42–51]. However it remains the problem that all the above models are unrealistic because of the assumptions made initially. A better approach has been made by models considering an RVE consisting of three phases i.e. the filler, the interphase and the matrix in the form of either concentric spheres in the case of particulates or in the form of coaxial cylinders for the fibre composites [52–59].

In the present paper a theoretical model is developed for the prediction of the elastic longitudinal modulus of elasticity E_{CL} of a unidirectional fibre reinforced composite. This model is consisted of three co-axial cylinders each one of them representing the fibre, the interphase and the matrix respectively. It is assumed that the fibre and matrix have well-defined properties while the interphase properties are varied along its thickness following an exponential law of variation. Moreover, the interphase thickness may be determined by means of thermal capacity measurements [23]. Two cases have been considered. In the first case a continuous variation of the elastic modulus, at the fibre–interphase common surface, was assumed while in the second case an abrupt variation in the elastic modulus at the same point has been considered. The results derived by considering these two cases were compared with other theories as well as with experimental results.

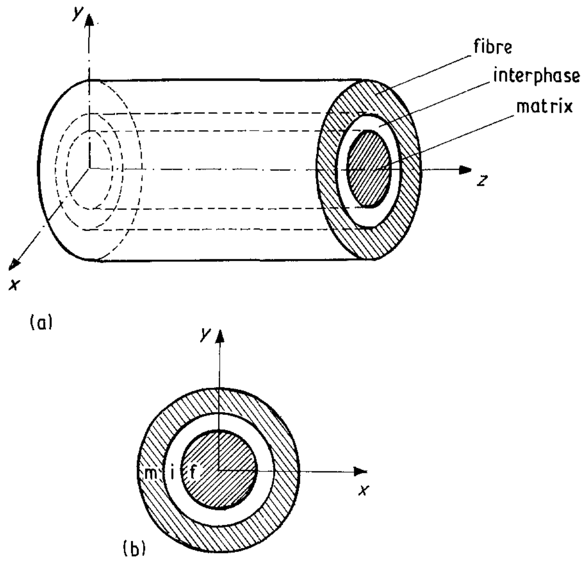

Figure 1 (a) Representative volume element (RVE) of the model considered and (b) its cross-sectional area.

2. The model

The transition from the microscopic level to the macroscopic one, i.e. to that of macroscopically or statistically homogeneous composite, is usually obtained through the concept of the representative volume element (RVE), i.e. a part of the composite sufficiently large to possess the same average properties as the bulk of the material.

Fig. 1a shows the representative volume element (RVE) of the model considered, whose cross-sectional area is shown in Fig. 1b.

If we denote by r_f, r_i, r_m the outer radii of the fibre, the interphase and the matrix circular sections respectively, then the fractions of the respective faces are given by

$$U_f = \frac{r_f^2}{r_m^2} \quad U_i = \frac{r_i^2 - r_f^2}{r_m^2} \quad U_m = \frac{r_m^2 - r_i^2}{r_m^2}$$

$$\text{with } U_m = (1 - U_f - U_i) \qquad (4)$$

where U denotes the volume fraction and the indices f, i and m correspond to the fibre, interphase and matrix respectively.

Following the same procedure described in a previous publication [58], the following expression for E_{CL} is obtained

$$\begin{aligned}\int_0^{r_m} E_{CL}\, \varepsilon^2 r\, dr = {} & \frac{1}{E_f}\int_0^{r_f} [8C_f^2(1 - v_f - 2v_f^2) \\ & + E_f^2\varepsilon^2] r\, dr + \int_{r_f}^{r_i} \frac{1}{E_i}\left(\frac{2A_i^2}{r^4}(1 + v_i)\right. \\ & \left. + 8C_i^2(1 - v_i - 2v_i^2) + E_i^2\varepsilon^2\right) r\, dr \\ & + \int_{r_i}^{r_m} \frac{1}{E_m}\left(\frac{2A_m^2}{r^4}(1 + v_m)\right. \\ & \left. + 8C_m^2(1 - v_m - 2v_m^2) + E_m^2\varepsilon^2\right) r\, dr \qquad (5)\end{aligned}$$

where: $\varepsilon = \varepsilon_{z,f} = \varepsilon_{z,m} = \varepsilon_{z,i}$ is the axial strain for each of the constituents of the composite, A, B, C are constants to be determined, v is Poisson's ratio and indices f, i, m state for the fibre, interphase and matrix respectively.

Next we define parameters α and β

$$\alpha = \frac{E_i(r_f)}{E_f} \quad \beta = \frac{v_i(r_f)}{v_f} \tag{6}$$

For $\alpha = \beta = 1$ a continuous variation of the elastic modulus and the Poisson's ratio is obtained at $r = r_f$. On the other hand, for $\alpha = E_m/E_f$ and $\beta = v_m/v_f$ the two-phase model (fibre–matrix) is obtained.

The following relationship between α and β is assumed to exist

$$\alpha = \frac{k}{\beta} + \lambda \tag{7}$$

where k, λ are constants which can be determined from the following conditions.

At $r = r_f$

$$\begin{aligned} &\text{for} \quad \alpha = 1 \Rightarrow \beta = 1 \\ &\text{for} \quad \alpha = E_m/E_f \Rightarrow \beta = v_m/v_f \end{aligned} \tag{8}$$

it follows that

$$k = \frac{1 - E_m/E_f}{1 - v_f/v_m} \quad \lambda = -\frac{v_f/v_m - E_m/E_f}{1 - v_f/v_m} \tag{9}$$

An exponential variation for $E_i(r)$ and $v_i(r)$ is considered

$$\begin{aligned} E_i(r) &= A_E \cdot r \cdot e^{-C_E \cdot r} + B_E \\ v_i(r) &= A_v \cdot r \cdot e^{-C_v \cdot r} + B_v \end{aligned} \tag{10}$$

where constants A_E, B_E, C_E and A_v, B_v, C_v can be evaluated from the boundary conditions:

(i) At $r = r_i$

$$\begin{aligned} E_i(r_i) &= E_m \quad v_i(r_i) = v_m \\ dE_i(r)/dr &= 0 \end{aligned} \tag{11}$$

where $R(r)$ is given by

$$R(r) = \frac{1 - r/r_i\, e^{(1-r/r_i)}}{1 - r_f/r_i\, e^{(1-r_f/r_i)}} \quad r_f \leqslant r \leqslant r_i \tag{14}$$

If we now introduce the parameter r' such as

$$r' = r/r_i \tag{15}$$

then Relation 13 may be written in the form

$$\begin{aligned} E_i(r') &= E_m + \left(\frac{(aE_f - E_m)}{(1 - U\, e^{(1-U)})}\right)(1 - r'\, e^{(1-r')}) \\ V_i(r') &= v_m + \left(\frac{(\beta v_f - v_m)}{(1 - U\, e^{(1-U)})}\right)(1 - r'\, e^{(1-r')}) \end{aligned} \tag{16}$$

where U is given by

$$U = \left(\frac{U_f}{U_i + U_f}\right)^{1/2} \quad U \leqslant r' \leqslant 1 \tag{17}$$

2.1. Determination of constants A_k, B_k, C_k (k = f, i, m)

The boundary conditions

$$\begin{aligned} E_i(r_i) &= E_m \quad v_i(r_i) = v_m \\ E_i(r_f) &= \alpha E_f \quad v_i(r_f) = \beta v_f \end{aligned} \tag{18}$$

are taken into account along with the conditions

$$\begin{aligned} &\text{At } r = r_f \quad \sigma_{r,f} = \sigma_{r,i} \\ &\text{At } r = r_i \quad \sigma_{r,i} = \sigma_{r,m} \\ &\text{At } r = r_f \quad u_{r,f} = u_{r,i} \\ &\text{At } r = r_i \quad u_{r,i} = u_{r,m} \\ &\text{At } r = r_m \quad \sigma_{r,m} = 0 \end{aligned} \tag{19}$$

we obtain

$$\begin{aligned} A_i = A_m &= F r_f^2 \quad C_f = \tfrac{1}{2}(1 - U_f)F \\ C_i = C_m &= -\tfrac{1}{2} U_f F \end{aligned} \tag{20}$$

where F is given by

$$F = \frac{aE_f v_f(1 - \beta)\varepsilon}{[a(1 - U_f)(1 - v_f - 2v_f^2) + U_f(1 - \beta v_f - 2\beta^2 v_f^2) + \beta v_f + 1]} \tag{21}$$

(ii) At $r = r_f$

$$E_i(r_f) = \alpha E_f \quad v_i(r_f) = \beta v_f$$

Thus the following results are obtained

$$\begin{aligned} A_E &= (aE_f - E_m)/(r_f\, e^{-(r_f/r_i)} - r_i\, e^{-1}) \\ A_v &= (\beta v_f - v_m)(r_f\, e^{-(r_f/r_i)} - r_i\, e^{-1}) \\ B_E &= E_m - c^{-1} \quad B_v = v_m - e^{-1} \\ C_E &= 1/r_i \quad C_v = 1/r_i \end{aligned} \tag{12}$$

Introducing now A_k, B_k, C_k ($k = E, v$) in Equation 10 we obtain for $E_i(r)$ and $v_i(r)$

$$\begin{aligned} E_i(r) &= E_m + (\alpha E_f - E_m)R(r) \\ v_i(r) &= v_m + (\beta v_f - v_m)R(r) \end{aligned} \tag{13}$$

E_{CL} can be evaluated by introducing these constants into Equation 5 and taking into account the final expression for $E_i(r)$ given by Equation 13.

For the special case where $\alpha = \beta = 1$, Relation 5 takes the simple form

$$E_{CL} = E_m U_m + E_f U_f + \frac{2}{r_m^2}\int_{r_f}^{r_i} E_i(r)\, r\, dr \tag{22}$$

3. Experimental procedure

The material used in the present study was a unidirectional glass fibre composite (Permaglass XE5/1, Permali Ltd., UK) consisting of a epoxy matrix and reinforced with long E-glass fibres. The matrix material was based on a diglycidyl ether of bisphenol A together with an aromatic amine hardener (Araldite MY 750/HT972, Ciba-Geigy, UK). The glass fibres

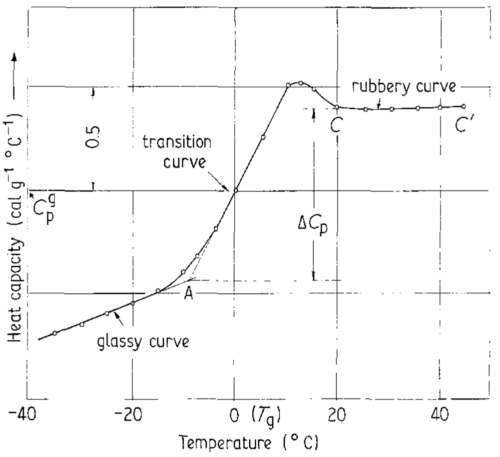

Figure 2 A typical DSC-trace for the specific heat capacity jump ΔC_p at the glass transition region of E-glass fibre epoxy composites and the mode of evaluation of ΔC_p.

had a diameter of 1.2×10^{-5} m and were contained at a volume fraction $U_f = 0.65$. The volume fraction U_f was determined, as customary, by igniting samples of the composite and weighting the residue, which gave the weight fraction of glass as $79.6 \pm 0.28\%$. This and the measured values of the relative densities of permaglass ($\varrho_g = 2.55$) and of the epoxy ($\varrho_e = 1.20$) gave the value $U_f = 0.65$. Complete descriptions of the material and its tensile creep properties are given by Theocaris and Papanicolaou [54, 60]. In order to determine the specific heat capacity values at the glass transition region, chip specimens with a 0.004 m diameter and thicknesses varying between 0.001 and 0.0015 m made either of the fibre composite with various values of U_f or of the matrix material, were tested on a differential scanning calorimetry (DSC) thermal analyser.

4. Evaluation of interphase characteristics

Fig. 2 shows a typical DSC trace for the specific heat capacity jump ΔC_p at the glass-transition region of E-glass fibre epoxy composites and the mode of evaluation of ΔC_p. Values from the values of ΔC_p^f and ΔC_p^0 the weight factor, λ, can be determined from Equation 2 and consequently the interphase thickness Δr can be found from Equation 3. Thus, since

$$r_i = r_f + \Delta r \quad \text{and} \quad U_i = \frac{r_i^2 - r_f^2}{r_m^2}$$

the respective values of U_i and r_i for the various fibre-volume contents can be determined. These values are given in Table I.

5. Variation of $E_i(r)$ – modulus

In a previous paper [58], four different laws of variation of $E_i(r)$ modulus, namely, a linear, a parabolic, an hyperbolic and a logarithmic, have been considered and their effect on the overall value of the longitudinal modulus of the composite E_{CL} had been investigated. In all cases it was assumed that $E_i(r)$, at $r = r_f$, equals E_f. However, this condition is hardly satisfied in real composites. Depending on the interfacial conditions one can expect an abrupt variation of the elastic modulus at the fibre–interphase common surface ($r = r_f$). This condition is fulfilled by the present model which takes into account this possibility through the parameter

$$\alpha = E_i(r_f)/E_f$$

Fig. 3 shows the mode of variation of $E_i(r)$ modulus as a function of the polar radius, r, according to the exponential law expressed by Equation 13 and for different values of the parameter α.

6. Effect of the mode of $E_i(r)$ – variation on E_{CL}

Values can be obtained by introducing the calculated values of $E_i(r)$ into Equation 5. Table II shows predicted E_{CL} values for different fibre volume fractions and for different α-values. From these values it becomes clear that E_{CL} values are insignificantly influenced by the parameter α. This result agrees well with the conclusion derived by Theocaris *et al.* [58] according to which the infinitesimal size of the extent of interphase, makes the modulus of the composite insensitive to the mode of variation of $E(r)$ modulus. *However, a strong effect of $E_i(r)$ mode of variation and especially of the elastic modulus abrupt variation of $r = r_f$ on the stress and strain fields around the fibre is expected. This effect will be the subject of future work.*

Predicted E_{CL} values derived from our model for the case $\alpha = \beta = 1$ were compared with respective values derived from our previous models [58] along with those derived from the unfolding model [61] as well as with experimental results from the literature [62, 63] and tabulated in Table I. It can be observed that these

TABLE I Predicted E_{CL} values derived from the present model for the case $\alpha = \beta = 1$ as compared with respective values from the literature

U_f	U_i ($\times 10^3$)	r_i (μm)	Mixture law	Parabolic law	Exponential law	Unfolding model	Reference [63]	Reference [62]
0.0	0	6.0	3.50	3.50	3.50	3.50	3.45	
0.10	1.2	6.036	10.35	10.38	9.90	10.36	10.41	
0.20	4.92	6.097	17.20	17.31	16.65	17.22	17.38	
0.30	11.09	6.110	24.05	24.30	23.48	24.07		
0.40	19.68	6.146	30.90	31.35	30.23	30.92		
0.50	30.75	6.182	37.75	38.45	37.27	37.77	38.28	
0.60	44.28	6.217	44.60	45.60	44.17	44.62	45.24	48.14 ± 0.82
0.65	52.0	6.235	48.03	49.13	47.65	48.13		52.15 ± 0.89
0.70	60.27	6.254	51.45	52.81	51.20	51.47		56.16 ± 0.96
0.80	78.72	6.288	58.30	60.08	58.27	58.31		
0.90	99.63	6.323	65.15	67.39	65.37	65.16		

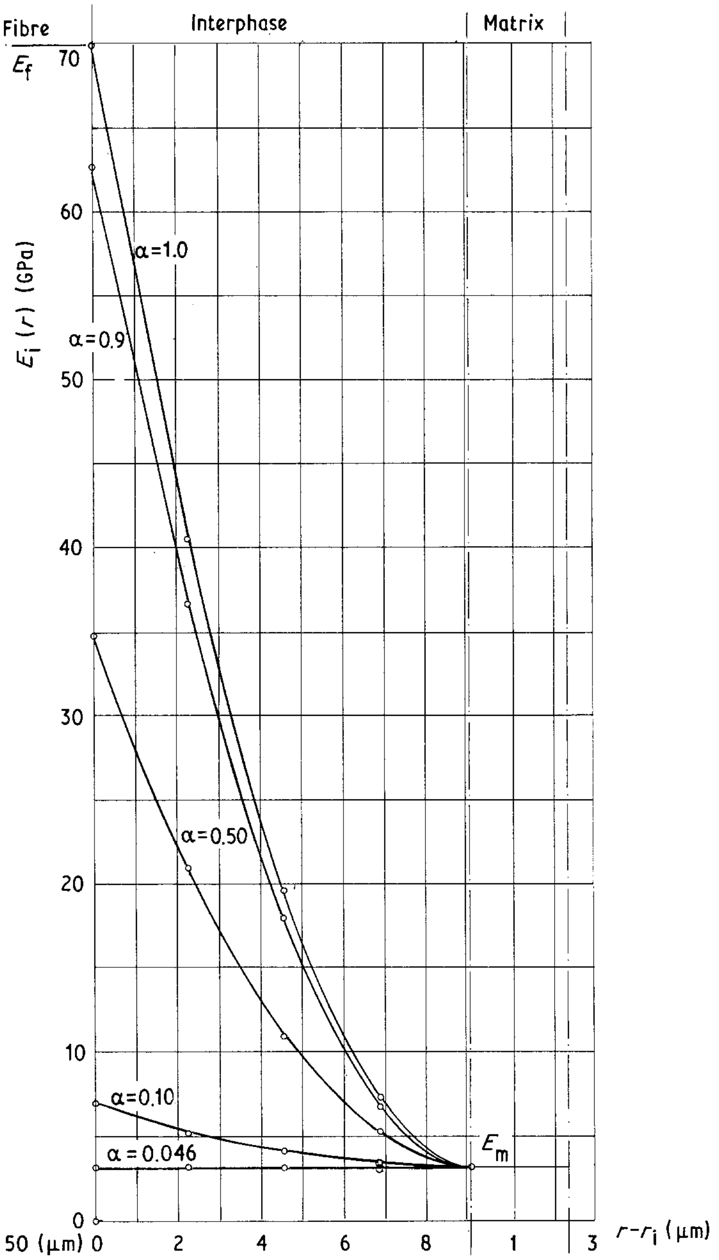

Figure 3 Mode of variation of $E_i(r)$ modulus as a function of the polar radius, r, according to the exponential law expressed by Equation 13 and for different values of the parameter α. $U_f = 0.50$.

values differ insignificantly and almost coincide with experimental values taken from the literature [62, 63].

Fig. 4 shows the variation of $E_i(r)$ modulus as a function of the polar radius r, according to different laws of variation. In the case of the exponential law of variation $\alpha = \beta = 1$ was taken in order for comparison to be made with the other models. It can be observed that values derived from the exponential law lie inbetween those derived by the parabolic law and those derived by the unfolding model.

Finally predicted E_{CL} values for $\alpha = \beta = 1$ are plotted against U_f in Fig. 5. In the same figure predicted values from other theories, as well as experimental values, are plotted for comparison. It can be seen that there is a good agreement between theory and experiment.

TABLE II Predicted E_{CL} values for different fibre volume fractions and for different α values

β	1.750	1.678	1.561	1.459	1.369	1.290	1.219	1.156	1.099	1.047	1.000
α	0.046	0.100	0.200	0.300	0.400	0.500	0.600	0.700	0.800	0.900	1.000
v_f											
0.10	9.89	9.94	10.05	10.12	10.14	10.12	10.07	10.01	9.95	9.91	9.90
0.20	16.57	16.66	16.85	16.98	17.03	17.00	16.92	16.82	16.73	16.66	16.65
0.30	23.25	23.37	23.61	23.79	23.86	23.84	23.75	23.64	23.53	23.46	23.45
0.40	29.92	30.06	30.35	30.56	30.66	30.65	30.57	30.47	30.37	30.31	30.31
0.50	36.59	36.74	37.05	37.29	37.41	37.43	37.38	37.31	37.23	37.20	37.23
0.60	43.25	43.41	43.72	43.98	44.13	44.19	44.18	44.15	44.12	44.13	44.19
0.65	46.59	46.74	47.05	47.31	47.47	47.56	47.58	47.58	47.58	47.61	47.69
0.70	49.92	50.07	50.38	50.63	50.81	50.92	50.97	51.00	51.04	51.10	51.21
0.80	56.58	56.73	57.01	57.26	57.46	57.62	57.75	57.87	57.98	58.12	58.28
0.90	63.24	63.38	63.62	63.86	64.09	64.31	64.52	64.74	64.95	65.17	65.40

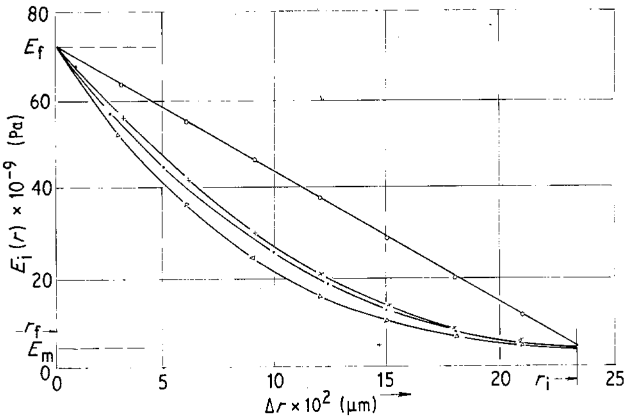

Figure 4 Variation of $E_i(r)$ modulus as a function of the polar radius r, according to different laws of variation. (x parabolic, ○ linear, △ unfolding, ● exponential)

7. Conclusions

A theoretical model has been developed for the prediction of the longitudinal modulus of elasticity, E_{CL}, of unidirectional fibre-reinforced composites. The existence of the interphase material has been taken into account and an exponential mode of variation of the interphase modulus $E_i(r)$ with the polar radius, r, was assumed. Moreover, the effect of an abrupt variation of the elastic modulus at $r = r_f$ on the overall longitudinal modulus of the composite was studied. It was found that

(i) Predicted E_{CL} values are insensitive to the mode of $E_i(r)$ variation as well as to the abrupt variation of the modulus at $r = r_f$.

(ii) Theoretical predictions agree well with both other theories and experimental results.

(iii) The discontinuity of the modulus at $r = r_f$ is expected to affect mainly the stress and strain fields around the inclusion.

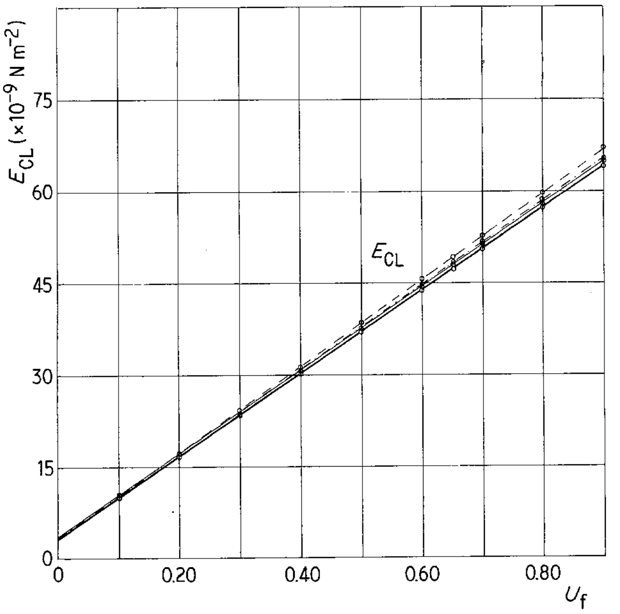

Figure 5 Variation of predicted E_{CL} values for $\alpha = \beta = 1$ against filler volume fraction U_f. (– – – parabolic, —·— [63], —— mixture law, —— exponential ($\alpha = \beta = 1$)).

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