

An upper bound of longitudinal elastic modulus for unidirectional fibrous composites as obtained from strength of materials approach

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Abstract

In this paper, an upper bound of the longitudinal elastic modulus of unidirectional fibrous composites is proposed according to strength of materials approach, on the premise that the fiber is much stiffer than the matrix. In the mathematical derivations, the concept of boundary interphase between fiber and matrix was also taken into account and the main objective of this work is the attainment of an upper bound for the interphase stiffness with respect to fiber concentration by volume. The novel element here is that the authors have not taken into consideration any specific variation law to approximate the interphase modulus. The theoretical results arising from the proposed formula were compared with those obtained from some reliable theoretical models as well as with experimental data found in the literature, and a satisfactory agreement was observed.

Keywords

fibrous composites; longitudinal modulus; upper bound; strength of materials; interphase

Introduction

The variational energy principles of classical elasticity theory were extensively used to determine upper and lower bounds on the moduli of unidirectional fiber – reinforced composites. For instance, Paul [1] used the principles of minimum energy and minimum complementary energy to define the bounds on the elastic modulus of a macroscopically isotropic, two – phase composite with arbitrary phase geometry. Yet, Hill [2] achieved to derive the same bounds via a different formalistic approach. Concurrently, Hashin and Rosen [3] constrained Paul's bounds in order to obtain a more useful evaluation of moduli for isotropic heterogeneous materials. However, approaches on the basis of energy principles generally result in bounds that may not be sufficiently close for practical use. A thorough and effective critique on theories predicting thermoelastic properties of fibrous composites was presented by Chamis and Sendeckyj [4]. Besides, a detailed survey, the aim of which was to review the analysis of composite materials from the applied mechanics and engineering viewpoint, was performed in Ref. [5]

On the other hand, a large amount of models towards the prediction of mechanical behavior of composites have a common characteristic of considering the fiber–matrix interface as a perfect mathematical surface. However, in reality the situation is much different mainly due to the roughness of the filler. Thus, around an inclusion embedded in a matrix a rather complex situation takes place, consisting of areas of imperfect bonding, permanent stresses resulting from shrinkage, and high – stress gradients or even stress singularities, attributed to the geometry of the inclusion, voids, microcracks etc. In addition, the interaction of the fiber with the matrix is usually a much more complicated procedure than a simple mechanical effect. The existence of a fiber actually restricts the segmental and molecular mobility of the polymeric matrix, as absorption interaction in polymer surface layers into fibers occurs. It is then evident that, under such circumstances, the quality of adhesion can hardly be quantified and a more thorough investigation by supposing the existence of an intermediate phase between matrix and filler is necessary. Indeed, the existence of a boundary interphase was experimentally verified by Lipatov [6] who estimated its thickness both for fibrous and particulate composites by means of Differential Scanning Calorimetry

(DSC) experiments. In this valuable work, it was also stated that the size of these heat capacity jumps for unfilled and filled materials is directly related to interphase thickness via empirical relationships.

Amongst a large number of theoretical models appeared in the literature, some of them take into account the existence this natural intermediate phase, developed during the preparation of the composite material and which plays the central role on its overall mechanical behaviour, as it characterises the effectiveness of the bonding between phases and defines an adhesion factor of the composite.

In a simplified approach which was adopted in Refs. [7, 8], this natural intermediate phase was assumed to be a homogeneous and isotropic material, whereas in Refs. [9, 10,11] more advanced and rigorous models were introduced, the main concept of which was the fiber to be occupied by a series of successive coaxial cylinders, each of which has a different elastic modulus in a step – function variation with respect to polar radius. In addition, valuable experimental investigations towards the estimation of mechanical and thermal properties of unidirectional fibrous composite materials were carried out by Clements and Moore [12] and Sih et al. [13]. In the meanwhile, an alternative viewpoint on the variable – modulus interphase concerning both particulate and fibrous composites is the two and three – term unfolding models introduced by Theocaris [14] which are based on the fact that the intermediate “phase” (termed mesophase annulus) constitutes a transition zone between fibers usually with high modulus and matrix usually with rather low modulus. In this context, the variable mesophase modulus is expressed with respect to the polar radius of an amended form of Hashin – Rosen cylinder assemblage model by relations of negative powers of the radius, which are compatible with their limits i.e. the moduli of fiber and matrix respectively. Further, Sideridis et al [15] proposed strength of materials and elasticity approaches to determine the elastic constants of fiber – reinforced composites, by taking into account the concept of boundary interphase. In this work, referring to longitudinal modulus the strength materials approach resulted in a modified form of standard mixtures law, whilst classical elasticity approach yielded an upper bound of this property. Also, the mode of variation of the variable interphase elastic properties was an n – th degree polynomial function with respect to interphase radius was initially considered, and for $n=2$ it yielded a parabolic law. However, given that the interphase zone is indeed a natural phase, (in particular a somewhat altered polymer matrix), and not an artificial one, in trying to cover the whole spectrum of the variation of its thermal and/or mechanical properties several laws have been adopted e.g. linear, parabolic, hyperbolic, logarithmic and exponential law [16]. In the past years, there is a lot of recent research work carried out for the determination of elastic constants of unidirectional fibrous composites and for the investigation of the effect of many parameters such as filler – matrix interaction, adhesion efficiency, fiber arrangement and vicinity etc. In Ref. [17] a micro – scale simulation and prediction of the mechanical properties of fibrous composites by means of the bridging micromechanics model was carried out, whilst for a thorough study on the effective properties of fibrous composite media of periodic structure, one may refer to Ref. [18]. Also, the effect of size and stacking of glass fibers on the mechanical properties of the fiber-reinforced-mortars was investigated in Ref. [19]. Finally, in Ref. [20] the influence of the statistical character of fiber strength on the predictability of tensile properties of polymer composites reinforced with natural filler was examined by comparing the well known linear and power – law Weibull models.

In the current work, the authors derived an upper bound of the longitudinal elastic modulus of unidirectional fibrous composites reinforced with continuous fibers according to strength of materials approach, on the premise that the fiber is much stiffer than the matrix. To estimate this bound the concept of an interphase layer developed between fiber and matrix having different properties was also taken into account and indeed the basic aim here is the achievement of an upper bound for the stiffness of this phase with respect to fiber content. The novelty of this investigation is that the authors have not considered any particular variation law to approximate the interphase modulus.

Towards the determination of an upper bound for longitudinal modulus

It is known that the use of the standard and/or inverse rule of mixtures to calculate the longitudinal and transverse modulus for a two – phase composite reinforced with unidirectional continuous fibers is based on the strength of materials approach and requires the following conditions to be fulfilled:

1. The fiber arrangement inside the matrix is uniform

2. The adhesion efficiency is perfect
3. Matrix is free of voids.
4. The applied loads are either parallel or perpendicular to the fiber direction.
5. The overall material is initially in a stress free condition, implying that no residual stresses appear.
6. Fiber and matrix behave as linearly elastic isotropic materials.

In Ref. [15] the concept of boundary interphase, was applied in the framework of a coaxial three – layer cylinder model, that constitutes a modified form of Hashin – Rosen cylinder assemblage model. Its cross – sectional area is exhibited in Fig. 1.

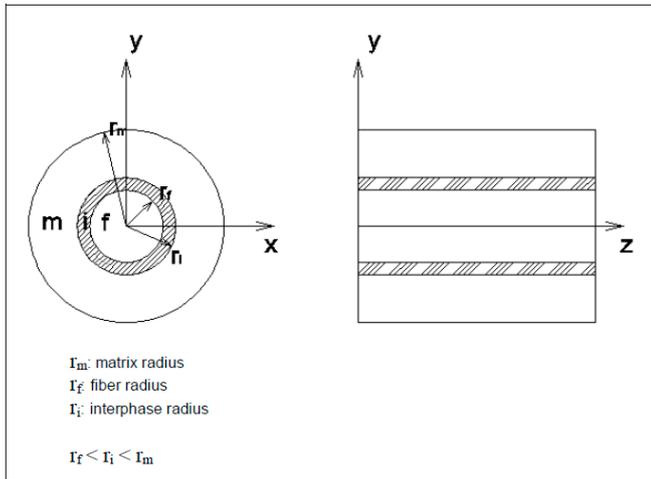


Fig. 1 Cross – sectional area and the three phase cylinder model

If we denote by $r_f; r_i; r_m$ the outer radii of the fiber, the interphase and the matrix circular sections respectively, then the volume fractions $U_f; U_i; U_m$ are given as

$$U_f = \frac{r_f^2}{r_m^2}; U_i = \frac{r_i^2 - r_f^2}{r_m^2}; U_m = \frac{r_m^2 - r_i^2}{r_m^2} \quad (1 \text{ a,b,c})$$

Apparently, as the filler volume fraction is increased the proportion of macromolecules characterized by a reduced mobility is also increased. This fact is synonymous with an augmentation in interphase concentration by volume. Lipatov [6] has shown that, if calorimetric measurements are performed in the neighborhood of the glass transition zone of the composite, energy jumps are observed. These jumps are too sensitive to the amount of filler added to the matrix and can be used to evaluate the boundary layers developed around the inclusions. This fact supports the empirical conclusion presented in Ref. [6], according to which the extent of the interphase expressed by its thickness Δr motivates the variation of the amplitudes of heat capacity jumps appearing at the glass transition zones of the matrix material and the composite with various filler – volume fractions. Moreover, the size of heat capacity jumps for unfilled and filled materials is directly related to Δr by an empirical relationship given in Ref. [6]. This expression defines the thickness Δr corresponding to the interphase and is written out below

$$\left(\frac{r_f + \Delta r}{r_f} \right)^2 - 1 = \frac{\lambda}{1 - U_f} \quad (2a)$$

where the coefficient λ is given by

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

Here, the numerator and the denominator of the fraction appearing in the right member of Eqn. (2b) are the sudden changes of the heat capacity for the filled and unfilled polymer respectively.

Next, to evaluate the volume fraction of the interphase layer, let us formulate eqn. (1b) as follows:

$$U_i = \frac{(r_f + \Delta r)^2 - r_f^2}{r_m^2} \Leftrightarrow$$

$$U_i = \frac{2r_f \Delta r + (\Delta r)^2}{r_m^2} \quad (3a)$$

Since $(\Delta r)^2 \ll r_m^2$ it implies that

$$\frac{(\Delta r)^2}{r_m^2} \cong 0 \quad (3b)$$

and therefore

$$U_i = \frac{2r_f \Delta r}{r_m^2} \quad (3c)$$

Eqn. (3c) can be combined with (1a) to yield

$$U_i = \frac{2U_f \Delta r}{r_f} \quad (4)$$

According to Strength of Materials approach, the longitudinal elastic modulus E_L for a fibrous composite material reinforced with unidirectional continuous fibers can be obtained from the following modified form of standard mixtures law, initially introduced in Ref. [15].

$$E_L = E_f U_f + E_m U_m + E_i U_i \quad (5)$$

Actually, the above relationship constitutes a “refined” expression of standard mixtures law for the apparent Young’s modulus in the direction of fibers.

Here, we emphasize that fiber, matrix and interphase which is somewhat an altered matrix, are isotropic. Now, as we have stated beforehand, in the current investigation we shall not taken into consideration any specific variation law to predict the interphase stiffness. On the contrary, the mathematical derivations resulting in an upper bound of the term E_i appearing in the above equation are based only on the assumption that the following inequality holds

$$E_m + E_i < E_f \quad (6)$$

Since the matrix is polymeric, this consideration does not generally contravene the generality.

Moreover, the following inequality is evident

$$E_m \leq E_i < E_f \quad (7)$$

Thus we can write out

$$\left\{ \begin{array}{l} E_m \leq E_i \\ \wedge \\ E_i \leq E_f \end{array} \right\} \Leftrightarrow \left\{ \begin{array}{l} 4\left(\frac{E_m}{2}\right)^2 \leq E_i^2 \\ \wedge \\ 4E_i^2 \leq 4E_i E_f \end{array} \right. \quad (8a,b)$$

and therefore

$$4\left(\frac{E_m}{2}\right)^2 + 4E_i^2 \leq E_i^2 + 4E_i E_f \Rightarrow E_i^2 \leq \frac{4}{3} \left[E_i E_f - \left(\frac{E_m}{2}\right)^2 \right] \Leftrightarrow$$

$$E_i^2 \leq \frac{4}{3} E_i E_f - \frac{E_m^2}{3} \quad (9)$$

In addition, on the basis of inequalities (6) and (7) one may deduce that the following inequalities also hold

$$\begin{aligned} (E_f - E_i) &\leq E_f + (E_f - E_m) \\ (E_f - E_m) &\leq (E_f - E_i) + E_f \\ E_f &\leq (E_f - E_m) + (E_f - E_i) \end{aligned} \quad (10 \text{ a,b,c})$$

Besides, the according to Fig. 1 the following inequality is plausible

$$r_f < r_i < r_m$$

and therefore

$$r_f^2 < r_i^2 < r_m^2 \quad (11a)$$

or equivalently

$$\frac{r_f^2}{r_m^2} < \frac{r_i^2}{r_m^2} < 1 \quad (11b)$$

Inequality (11b) by the aid of eqns. (1 a,b,c) yields

$$U_f < U_i + U_f < 1 \quad (11c)$$

Now let us set

$$1 = x; U_i + U_f = y; U_f = z \quad (12a,b,c)$$

where the auxiliary variables $x; y; z$ such that $x > y > z$ lie in the interval $[0,1]$

Next, one may observe that the following inequality holds identically

$$[(E_f - E_i)(x - y) - E_f(y - z)]^2 \geq 0 \quad (13)$$

and therefore

$$\begin{aligned} (E_f - E_i)^2(x - y)^2 + E_f^2(y - z)^2 - 2(E_f - E_i)E_f(x - y)(y - z) &\geq 0 \Leftrightarrow \\ (E_f - E_i)^2(x - y)[(x - z) - (y - z)] + E_f^2(y - z)[-(x - y) - (z - x)] - 2(E_f - E_i)E_f(x - y)(y - z) &\geq 0 \Leftrightarrow \\ (E_f - E_i)^2(x - y)(x - z) - E_f^2(x - y)(y - z) - (E_f - E_i)^2(x - y)(y - z) - 2(E_f - E_i)E_f(x - y)(y - z) + E_f^2(z - x)(z - y) &\geq 0 \Leftrightarrow \\ (E_f - E_i)^2(x - y)(x - z) - [E_f^2 + (E_f - E_i)^2 + 2(E_f - E_i)E_f](x - y)(y - z) + E_f^2(z - x)(z - y) &\geq 0 \Leftrightarrow \\ (E_f - E_i)^2(x - y)(x - z) - [E_f + (E_f - E_i)]^2(x - y)(y - z) + E_f^2(z - x)(z - y) &\geq 0 \quad (14) \end{aligned}$$

Concurrently, according to inequality (10b) we infer

$$(E_f - E_m)^2 \leq [E_f + (E_f - E_i)]^2 \Leftrightarrow -(E_f - E_m)^2 \geq -[E_f + (E_f - E_i)]^2 \quad (15)$$

Then given that the terms $(x - y); (y - z)$ agree in sign, inequality (14) can be combined with (15) to yield

$$\begin{aligned} (E_f - E_i)^2(x - y)(x - z) - (E_f - E_m)^2(x - y)(y - z) + E_f^2(z - x)(z - y) &\geq 0 \Leftrightarrow \\ (E_f - E_i)^2(x - y)(x - z) + (E_f - E_m)^2(y - x)(y - z) + E_f^2(z - x)(z - y) &\geq 0 \Leftrightarrow \\ E_f^2(x - y)(x - z) + E_i^2(x - y)(x - z) + (E_f - E_m)^2(y - x)(y - z) + E_f^2(z - x)(z - y) &\geq 2E_f E_i(x - y)(x - z) \end{aligned} \quad (16)$$

Finally since the terms $(x - y); (x - z)$ agree in sign, inequality (16) can be combined with (9) to yield

$$\begin{aligned} E_f^2(x - y)(x - z) + \left(\frac{4E_i E_f}{3} - \frac{E_m^2}{3} \right) (x - y)(x - z) + (E_f - E_m)^2(y - x)(y - z) + E_f^2(z - x)(z - y) &\geq 2E_f E_i(x - y)(x - z) \Leftrightarrow \\ E_f^2(x - y)(x - z) - \frac{E_m^2}{3}(x - y)(x - z) + (E_f - E_m)^2(y - x)(y - z) + E_f^2(z - x)(z - y) &\geq \left(2 - \frac{4}{3} \right) (x - y)(x - z) E_f E_i \Leftrightarrow \\ E_i &\leq \frac{E_f^2(x - y)(x - z) - \frac{E_m^2}{3}(x - y)(x - z) + (E_f - E_m)^2(y - x)(y - z) + E_f^2(z - x)(z - y)}{\frac{2}{3} E_f(x - y)(x - z)} \end{aligned} \quad (17)$$

Since we have set $1 = x; U_i + U_f = y; U_f = z$ it follows that

$$E_i \leq \frac{E_f^2(1 - U_i - U_f)(1 - U_f) - \frac{E_m^2}{3}(1 - U_i - U_f)(1 - U_f) + (E_f - E_m)^2(U_i + U_f - 1)U_i + E_f^2(1 - U_f)U_i}{\frac{2}{3} E_f(1 - U_i - U_f)(1 - U_f)} \quad (18)$$

Thus the above inequality designates an upper bound for the interphase stiffness which results from the following relationship

$$\max E_i = \frac{\left(E_f^2 - \frac{E_m^2}{3} \right) U_m(1 - U_f) + E_f^2(1 - U_f)U_i - (E_f - E_m)^2 U_m U_i}{\frac{2}{3} E_f U_m(1 - U_f)} \quad (19)$$

Here one may pinpoint that the above relationship is independent of any variation law that could be adopted to predict in an exact manner how the interphase modulus varies with radius of the coaxial cylinder model. We emphasize that the interphase zone is a natural phase which is developed in reality between filler and polymer matrix and is neither an artificial one, e.g. created by the immersion of the fibers in an agent, nor a pseudophase being contrived to simulate the microstructure of the composite.

Now one may observe that according to the proposed technique the following three intermediate steps relating to the process of determining the stiffness of the overall composite material are bypassed:

- i) Approximation of the stiffness of interphase layer by a polynomial function or any other arbitrary continuous function with respect to the radius of the coaxial cylindrical three layer model.
- ii) Estimation of the average values of stiffness for the interphase zone. This procedure takes place to accommodate the calculations, as it can be observed in Ref. [15].

iii) Measurement of the thickness of interphase zone by means of DSC experiments.

Yet one may point out that it has been proved [21, 22] that for fibrous composites reinforced either with long or with short fibers the ordered pairs (U_i, U_f) fit in an excellent manner a two degree parabola expressed by the following relation

$$U_i = 0.123 \cdot U_f \quad (20)$$

It can therefore be noticed that the process of calculating the longitudinal modulus of the final material has shortened considerably, when compared with the corresponding procedures presented in Refs. [11, 14, 15, 16].

In his context, one may observe that via this performed analytical technique, the upper bound of the interphase stiffness has been represented directly with respect to filler content without having previously been expressed as a single – valued function of the polar radius of the coaxial three phase cylinder model. Then eqn. (5) can be combined with (19) to yield an upper bound for the longitudinal modulus of the overall material in the following explicit form

$$\max E_L = E_f U_f + E_m U_m + \frac{\left[\left(E_f^2 - \frac{E_m^2}{3} \right) U_m (1 - U_f) + E_f^2 (1 - U_f) U_i - (E_f - E_m)^2 U_m U_i \right] \cdot U_i}{\frac{2}{3} E_f U_m (1 - U_f)} \quad (21)$$

Thus, eqn. (21) in association with the eqn. (20) leads to the direct calculation of an upper bound for the longitudinal composite modulus in terms of fiber volume fraction.

Theoretical formulae used for comparison

Now, let us make the following brief theoretical remarks referring to some reliable theoretical formulae resulting in the direct estimation of longitudinal modulus for a general class of unidirectional fibrous composites reinforced with continuous fibers.

a) Theocaris Sideridis and Papanicolaou formula [Ref. 11]

The adopted microstructural model is a coaxial three layer cylindrical model similar to that presented in Fig. 1. The longitudinal modulus E_L , was calculated in the framework of energy balance for the overall material, and therefore

$$\begin{aligned} \frac{1}{2} \int_{V_c} E_L \varepsilon^2 dV_c &= \frac{1}{2} \int_{V_f} (\sigma_{r,f} \varepsilon_{r,f} + \sigma_{\theta,f} \varepsilon_{\theta,f} + \sigma_{z,f} \varepsilon_{z,f}) dV_f + \\ &+ \frac{1}{2} \int_{V_i} (\sigma_{r,i} \varepsilon_{r,i} + \sigma_{\theta,i} \varepsilon_{\theta,i} + \sigma_{z,i} \varepsilon_{z,i}) dV_i + \\ &+ \frac{1}{2} \int_{V_m} (\sigma_{r,m} \varepsilon_{r,m} + \sigma_{\theta,m} \varepsilon_{\theta,m} + \sigma_{z,m} \varepsilon_{z,m}) dV_m \end{aligned} \quad (22)$$

or equivalently

$$\begin{aligned}
\frac{1}{2} \int_0^{r_m} E_L \varepsilon^2 2\pi r h dr &= \frac{1}{2} \int_0^{r_m} (\sigma_{r,f} \varepsilon_{r,f} + \sigma_{\theta,f} \varepsilon_{\theta,f} + \sigma_{z,f} \varepsilon_{z,f}) 2\pi r h dr + \\
&+ \frac{1}{2} \int_{r_f}^{r_i} (\sigma_{r,i} \varepsilon_{r,i} + \sigma_{\theta,i} \varepsilon_{\theta,i} + \sigma_{z,i} \varepsilon_{z,i}) 2\pi r h dr + \\
&+ \frac{1}{2} \int_{r_i}^{r_m} (\sigma_{r,m} \varepsilon_{r,m} + \sigma_{\theta,m} \varepsilon_{\theta,m} + \sigma_{z,m} \varepsilon_{z,m}) 2\pi r h dr
\end{aligned} \tag{23}$$

The strains for each phase are given as

$$\begin{aligned}
\varepsilon_{r,f} &= \frac{du_{r,f}}{dr} = \frac{1}{E_f} \left[2C(1 - \nu_f - 2\nu_f^2) - \nu_f E_f \varepsilon \right] \\
\varepsilon_{\theta,f} &= \frac{u_{r,f}}{r} = \frac{1}{E_f} \left[2C(1 - \nu_f - 2\nu_f^2) - \nu_f E_f \varepsilon \right] \\
\varepsilon_{r,i} &= \frac{du_{r,i}}{dr} = \frac{1}{E_i} \left[(1 + \nu_i) \frac{K}{r^2} + 2M(1 - \nu_i - 2\nu_i^2) - \nu_i E_i \varepsilon \right] \\
\varepsilon_{\theta,i} &= \frac{u_{r,i}}{r} = \frac{1}{E_i} \left[-(1 + \nu_i) \frac{K}{r^2} + 2M(1 - \nu_i - 2\nu_i^2) - \nu_i E_i \varepsilon \right] \\
\varepsilon_{r,m} &= \frac{du_{r,m}}{dr} = \frac{1}{E_m} \left[(1 + \nu_m) \frac{F}{r^2} + 2H(1 - \nu_m - 2\nu_m^2) - \nu_m E_m \varepsilon \right] \\
\varepsilon_{\theta,m} &= \frac{u_{r,m}}{r} = \frac{1}{E_m} \left[-(1 + \nu_m) \frac{F}{r^2} + 2H(1 - \nu_m - 2\nu_m^2) - \nu_m E_m \varepsilon \right]
\end{aligned} \tag{24 a,b,c,d,e,f}$$

Hence it follows

$$\begin{aligned}
\int_0^{r_m} E_L \varepsilon^2 r dr &= \int_0^r \left\{ \frac{1}{E_f} \left[8C^2(1 - \nu_f - 2\nu_f^2) + E_f^2 \varepsilon^2 \right] \right\} r dr + \\
&+ \int_{r_f}^{r_i} \left\{ \frac{1}{E_i} \left[2(1 + \nu_i) \frac{K^2}{r^4} + 8M^2(1 - \nu_i - 2\nu_i^2) + E_i^2 \varepsilon^2 \right] \right\} r dr + \\
&+ \int_{r_i}^{r_m} \left\{ \frac{1}{E_m} \left[2(1 + \nu_m) \frac{F^2}{r^4} + 8H^2(1 - \nu_m - 2\nu_m^2) + E_m^2 \varepsilon^2 \right] \right\} r dr
\end{aligned} \tag{25}$$

and therefore

$$\int_0^{r_m} E_L \varepsilon^2 r dr = \int_0^{r_i} \left[8C^2(1 - \nu_f - 2\nu_f^2) + E_f^2 \varepsilon^2 \right] r dr + \int_{r_i}^{r_i} \frac{1}{E_i} \left[\frac{2K^2}{r^4} (1 + \nu_i) + 8M^2(1 - \nu_i - 2\nu_i^2) + E_f^2 \varepsilon^2 \right] r dr + \frac{1}{E_m} \int_{r_i}^{r_m} \left[\frac{2F^2}{r^4} (1 + \nu_m) + 8H^2(1 - \nu_m - 2\nu_m^2) + E_m^2 \varepsilon^2 \right] r dr \quad (26)$$

In the above relation both interphase modulus and the Poisson ratio are functions of the distance from the fiber according to a three phase cylinder model. To integrate the second term four different laws of variation were taken into account.

Hence the above relation according these variation laws yields the following explicit expressions

a) Linear Law for $E_i(r)$ and $\nu_i(r)$

$$E_i(r) = A + Br \quad \text{and} \quad \nu_i(r) = A' + B'r \quad \text{with} \quad r_f \leq r \leq r_i \quad (27a,b)$$

and therefore

$$E_i(r) = \frac{E_f r_i - E_m r_f}{r_i - r_f} - \frac{E_f - E_m}{r_i - r_f} r \quad (28a,b)$$

$$\nu_i(r) = \frac{\nu_f r_i - \nu_m r_f}{r_i - r_f} + \frac{\nu_m - \nu_f}{r_i - r_f} r$$

b) Hyperbolic Law

$$E_i(r) = A + B/r \quad \text{and} \quad \nu_i(r) = A' + B'/r \quad \text{with} \quad r_f \leq r \leq r_i \quad (29 a,b)$$

thus it implies that

$$E_i(r) = \frac{E_m r_i - E_f r_f}{r_i - r_f} + \frac{(E_f - E_m) r_f r_i}{(r_i - r_f) r}, \quad \nu_i(r) = \frac{\nu_m r_i - \nu_f r_f}{r_i - r_f} + \frac{(\nu_f - \nu_m) r_f r_i}{(r_i - r_f) r} \quad (30a,b)$$

c) Parabolic law

$$E_i(r) = Ar^2 + Br + C \quad \text{and} \quad \nu_i(r) = A'r^2 + B'r + C'r \quad \text{with} \quad r_f \leq r \leq r_i \quad (31a,b)$$

To estimate A, B, C and A', B', C' one may consider that $E_i(r)$ is minimum whilst $v_i(r)$ is maximum at $r = r_i$.

$$r = r_i : \frac{dE_i(r)}{dr} = 0 \text{ with } \frac{d^2E_i(r)}{dr^2} > 0 \text{ and } \frac{dv_i(r)}{dr} = 0 \text{ with } \frac{d^2v_i(r)}{dr^2} < 0$$

$$E_i(r) = \frac{(E_f - E_m)r^2 - 2(E_f - E_m)r_i r + E_f r_i^2 + E_m r_f^2 - 2E_m r_f r_i}{(r_i - r_f)^2}$$

$$v_i(r) = \frac{(v_f - v_m)r^2 - 2(v_m - v_f)r_i r + v_f r_i^2 + v_m r_f^2 - 2v_m r_f r_i}{(r_i - r_f)^2} \quad (32 \text{ a,b})$$

Hence the longitudinal modulus of the composite for each variation law is obtained from the following formulae

i) For linear law

$$E_L = E_f \frac{r_f^2}{r_m^2} + \frac{(E_f r_i - E_m r_f)}{r_m^2} (r_i + r_f) - \frac{2(r_i^2 + r_i r_f + r_f^2)(E_f - E_m)}{3r_m^2} +$$

$$+ \left[\frac{1 - v_m - 2v_m^2}{E_m} + E_m \right] \frac{(r_m^2 - r_i^2)}{r_m^2} \quad (33)$$

Here it was taken into account that according to Lipatov method [6] the following equality holds

$$\frac{r_f^2}{r_i^2} = \frac{U_f}{U_f + U_i} = \frac{1 - U_f}{1 - U_f(1 - \lambda)} \quad (34)$$

thus it follows

$$E_L = E_f U_f + \frac{1}{3} \left[(E_f + 2E_m)(1 - U_m) - (2E_f + E_m)U_f + (E_f - E_m)\sqrt{U_f(1 - U_m)} \right] +$$

$$+ \left[\frac{1 - v_m - 2v_m^2}{E_m} + E_m \right] U_m \quad (35)$$

ii) For hyperbolic law

$$E_L = E_f \frac{r_f^2}{r_m^2} + E_m \left(\frac{r_m^2 - r_i^2}{r_m^2} \right) + \frac{E_m r_i - E_f r_f}{r_m^2} (r_f + r_i) + 2(E_f - E_m) \frac{r_f r_i}{r_m^2} + \left[\frac{1 - v_m - 2v_m^2}{E_m} + E_m \right] \left(\frac{r_m^2 - r_i^2}{r_m^2} \right) \quad (36)$$

Then, by the aid of eqn. (34) it implies that

$$E_L = E_f U_f + (E_m \sqrt{1-U_m} - E_f \sqrt{U_f}) (\sqrt{1-U_m} + \sqrt{U_f}) + 2(E_f - E_m) (\sqrt{U_f(1-U_m)}) + \left[\frac{1-\nu_m - 2\nu_m^2}{E_m} + E_m \right] U_m \quad (37)$$

iii) For parabolic law

$$E_L = E_f \frac{r_f^2}{r_m^2} + \frac{(E_f - E_m)(r_i^3 + r_i^2 r_f + r_i r_f^2 + r_f^3)}{2r_m^2(r_i - r_f)} - \frac{4(E_f - E_m)r_i(r_i^2 + r_f r_i + r_f^2)}{r_m^2(r_i - r_f)} + \frac{(E_f r_i^2 + E_m r_f^2 - 2E_m r_f r_m)(r_f + r_i)}{r_m^2(r_i - r_f)} + \left[\frac{1-\nu_m - 2\nu_m^2}{E_m} + E_m \right] \left(\frac{r_m^2 - r_i^2}{r_m^2} \right) \quad (38)$$

and by the aid of eqn. (34) it implies that

$$E_L = E_f U_f + E_m U_m + \frac{3(E_f - E_m) \left(\sqrt{(1-U_m)^3} + \sqrt{U_f(1-U_m)} + \nu_f \sqrt{1-U_m} + \sqrt{U_f^3} \right)}{6(\sqrt{1-U_m} - \sqrt{U_f})} - \frac{8(E_f - E_m) \left(\sqrt{1-U_m} \right) (1-U_m + U_f + \sqrt{U_f(1-U_m)})}{6(\sqrt{1-U_m} - \sqrt{U_f})} + \left[\frac{1-\nu_m - 2\nu_m^2}{E_m} + E_m \right] U_m \quad (39)$$

b) Theocaris' Two – Term Unfolding Model for Fibrous Composites [Ref. 14]

$$E_{LC} = E_m + E_f U_f - \frac{E_m U_f}{3} + \frac{E_f U_f}{\eta - 1} (1 - B^{\eta-1}) + \frac{E_f U_f B^\eta}{3} \left(1 + \frac{1}{\sqrt{B}} - \frac{2}{B} \right) - \frac{E_m U_f}{3} \left(\frac{1}{\sqrt{B}} + \frac{1}{B} \right) \quad (40)$$

where the coefficient η , was experimentally determined in Ref. [14] and also the real quantity B depends on the fiber volume fraction and the coefficient λ and signifies the implicit role of the mesophase annulus in this formula.

c) Theocaris' Three – Term Unfolding Model for Fibrous Composites [Ref. 14]

$$E_{LC} = E_m + E_f U_f - E_m U_f + U_f \left[\frac{E_f}{\frac{\eta}{2} - 1} \left(1 - B^{\frac{\eta}{2}-1} \right) - \frac{E_m}{\eta - 1} (1 - B^{\eta-1}) \right] \quad (41)$$

Again, the quantities η and B are determined in the same way as in Two – Term Unfolding Model.

d) Sideridis – Papadopoulos - Kyriazi formula [Ref. 15]

According to a classical elasticity approach an upper bound for longitudinal modulus is given by the following explicit expression

$$E_L = \frac{(1-\nu_f - 4\nu\nu_f + 2\nu^2)}{(1-\nu_f - 2\nu_f^2)} E_f U_f + \frac{(1-\nu_i - 4\nu\nu_i + 2\nu^2)}{(1-\nu_i - 2\nu_i^2)} E_i U_i + \frac{(1-\nu_m - 4\nu\nu_m + 2\nu^2)}{(1-\nu_m - 2\nu_m^2)} E_m U_m \quad (42)$$

with

$$v_L = \frac{(1-v_f - 2v_f^2)E_f U_f v_f + (1-v_i - 2v_i^2)E_i U_i v_i + (1-v_m - 2v_m^2)E_m U_m v_m}{(1-v_f - 2v_f^2)E_f U_f + (1-v_i - 2v_i^2)E_i U_i + (1-v_m - 2v_m^2)E_m U_m} \quad (43)$$

Again, the Poisson ratio of interphase zone can be evaluated for linear, hyperbolic and parabolic variation respectively via the same formulae that were introduced in Ref. [11] and we previously remarked i.e. eqns. (28b, 30b, 32b).

Results and Discussion

In Table 1 the theoretical predictions concerning the upper bound of the composite longitudinal modulus, as obtained from eqn. (21) in combination with eqn. (20) appear with respect to fiber content which is up to 0.65. Roughly speaking one may notice that this value generally constitutes the optimum fiber volume fraction above which the reinforcing action of the fibers is upset. For facility reasons and to be in accordance with the experimental works of Clements and Moore [12] and Sih et al [13], we considered an epoxy composite reinforced with E – glass fibers, i.e. $E_f = 70 \text{ GN/m}^2$ and $E_m = 3.5 \text{ GN/m}^2$ whereas $v_f = 0.2$ and $v_m = 0.35$.

In the same table, the theoretical values obtained from Theocaris et al [11], eqn. (22), according to three variation laws also occur. Besides, the theoretical values yielded by two and three term unfolding models derived from Theocaris [14] are presented. Finally, the theoretical values achieved by the expression for apparent Young's modulus in the direction of fibers obtained from Sideridis et al [15] according to three variation laws are exhibited. On the other hand, the experimental values obtained from Clements and Moore [12] and Sih et al [13] are performed as well.

Here, one may observe that amongst the variation laws that were adopted by Theocaris et al. and Sideridis et al. to approach the interphase stiffness in the formulae presented in Ref. [11] and Ref. [15] respectively, i.e. eqn. (26) and (42) in the previous Section, the linear ones yield the highest predictions.

U_f	E_L GN/m ²	authors	Theocaris et al [11] Linear Law	Theocaris et al [11] Hyperbolic Law	Theocaris et al [11] parabolic Law	Sideridis et al [15] Linear Law	Sideridis et al [15] Hyperbolic Law	Sideridis et al [15] Parabolic Law	Two Term Unfolding model [14]	Three Term Unfolding model [14]	Clements and Moore [12]	Sih et al [13]
0		3.5	3.5	3.5	3.5	3.5	3.5	3.15	3.5	3.5		3.45
0.1		11.3634	10.39	10.39	10.38	11.0134	10.9095	10.7952	9.8913	10.3262		10.41
0.2		19.2330	17.37	17.37	17.31	18.4122	18.2385	18.0024	16.7535	17.2326		17.38
0.3		27.1150	24.43	24.43	24.30	25.8958	25.6515	25.272	22.3868	23.2757		
0.4		35.0231	31.57	31.57	21.35	33.4642	33.1485	22.204	27.1696	28.1174		
0.5		42.9913	38.80	38.79	38.45	41.128	40.7295	39.988	32.7749	34.8412		
0.6		51.1192	46.11	46.09	45.60	48.8766	48.3945	47.424	38.5921	40.6587	38.28	48.14
0.65		55.3338	49.75	49.73	49.13	52.735	52.2165	51.0952	43.3574	46.2139	45.24	52.15

Table 1 Theoretical and experimental values of longitudinal modulus with respect to fiber concentration

Next, Fig. 2 designates the theoretical values for the upper bound of the composite longitudinal modulus, yielded by eqn. (21) associated with eqn. (20) versus fiber volume fraction, together with those obtained from Theocaris et al. (linear variation law) [11] and Sideridis et al. (linear variation law) [15]. Moreover, the theoretical predictions given by Theocaris' two and three term unfolding model [14] are also performed.

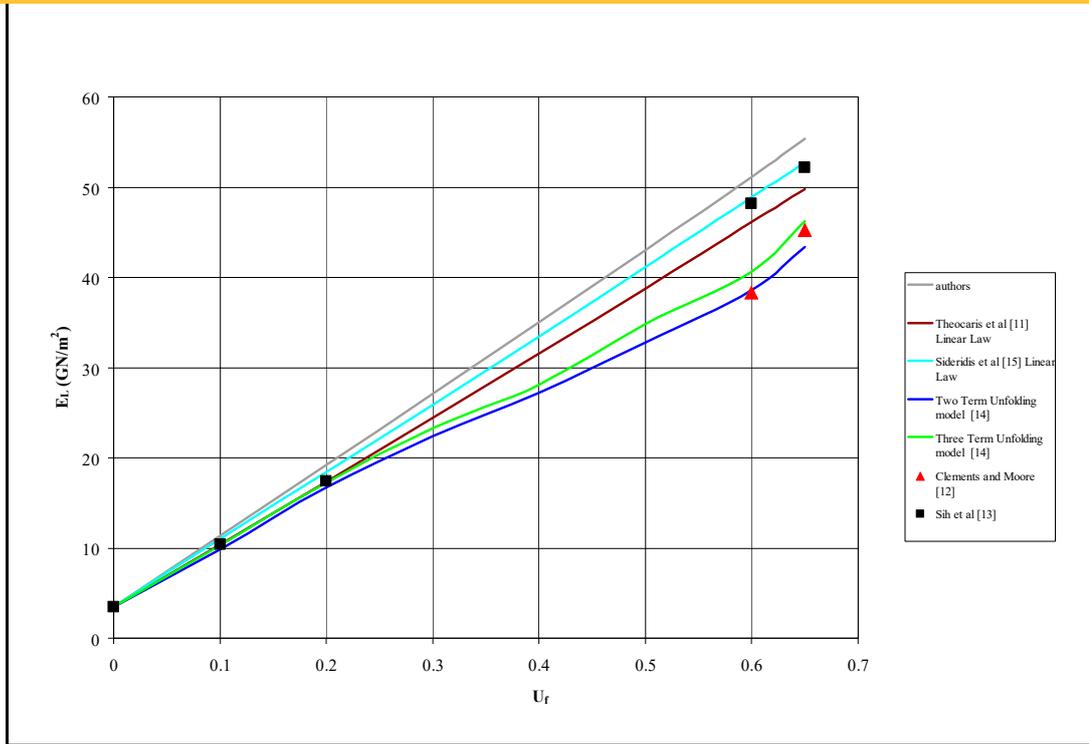


Fig. 2 Variation of Longitudinal modulus against fiber volume fraction

By focusing in Fig. 2, one may observe that for low filler contents the theoretical values of E_L arising from eqn. (21) by the aid of (20) are close to those yielded by all the other theoretical formulae used for comparison. In addition, they are in good agreement with the experimental values obtained from Sih et al [13]. Next, for medium fiber concentrations by volume one may point out a deviation especially between the values obtained from eqn. (21) as well as from Sideridis' upper bound [15] i.e. eqn. (42) of previous section, with the predictions given by the other three theoretical formula [11,14] and experimental results. An increment of this deviation is noticed at high filler contents up to 0.65. Besides, the theoretical predictions of eqn. (21) are above the experimental data obtained from Sih et al. [13] and well above those of Clements and Moore [12]. Of course one may elucidate that such discrepancies could be generally expected given that the aim of eqns. (21) and (42) as well, is to signify an upper bound of the longitudinal modulus. Moreover, some discrepancies are also attributed to the fact that some theoretical assumptions and conceptions cannot be fulfilled in praxis.

In the meanwhile, one may also observe that for high fiber contents the theoretical values yielded by Theocaris' two and three term unfolding model [14] are in consensus with the experimental results obtained from Clements and Moore [12]. Finally, it can be said that eqn. (21) definitely constitutes an improvement when compared with Sideridis' upper bound [15].

Nevertheless, a shortcoming of eqn. (19) which illustrates the maximum value of interphase stiffness is that it was based on the hypothesis that the fiber is much stiffer than the matrix i.e. $E_m + E_i < E_f$ and hence an analogous expression for Poisson ratio of interphase cannot be derived according to this reasoning. Thus the overall methodology leading to an upper bound of interphase modulus without the consideration of any variation law, cannot be used to improve theoretical formulae arising from elasticity approach, such as eqn. (26) and eqn. (42) of previous Section, without the adoption of several variation laws to approach interphase Poisson ratio. Thus, one could not know beforehand which variation law of interphase Poisson ratio would yield the highest predictions of E_L in the final expression of longitudinal modulus.

On the other hand one may also pinpoint that since according to strength of materials approach fibers, matrix and interphase are assumed somewhat *as solid blocks with volumes analogous to their relative abundance in the entire material* instead of the modified form of Hashin – Rosen cylinder assemblage model presented in Fig. 1 one may adopt the following simplified model to simulate the microstructure of the unidirectional fibrous composite.

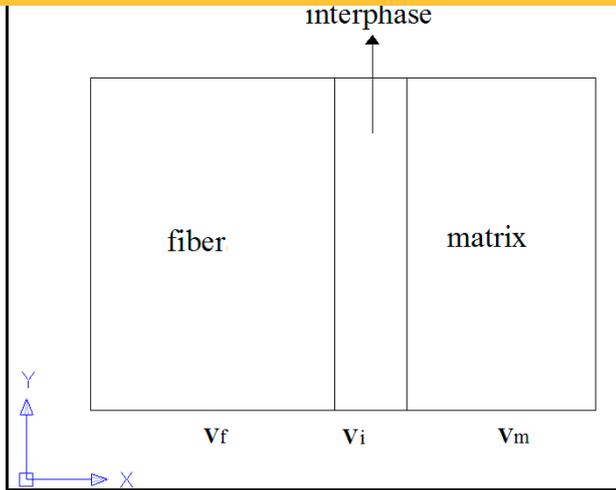


Fig. 3 Simplified model of the composite structure

Evidently the following expressions hold

$$U_m = \frac{V_m}{V}; U_f = \frac{V_f}{V}; U_i = \frac{V_i}{V} \quad (44)$$

with $V = V_m + V_f + V_i$

Obviously, since the inequality $U_f < U_i + U_f < 1$ holds, the overall mathematical procedure resulting in eqn. (19) and in eqn. (21) remains as is.

Conclusions

In this theoretical work, an upper bound of longitudinal modulus of three phase fibrous – reinforced composites was determined according to strength of materials approach, in terms of the constituent material properties, on the presumption that the fiber is much stiffer than the matrix, fact that generally concerns polymeric composites. The fibers of the overall material, which is supposed to be of periodic microstructure, are unidirectional continuous and isotropic. Concurrently, the concept of interphase in the context of a cluster of three coaxial cylinders unit cell was taken into consideration. The novel element here is that the authors did not assume any specific variation law to approximate the interphase modulus. In this context, three basic intermediate steps referring to the process of estimating the stiffness of the overall material, i.e. theoretical approximation of interphase stiffness, estimation of its average values and measurement interphase thickness via DSC experiments, were bypassed.

The theoretical predictions obtained from the proposed formula were compared with several theoretical models together with experimental data found in the literature, and a reasonable agreement was observed.

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