

Research Article

A Theoretical Consideration on the Estimation of Interphase Poisson's Ratio for Fibrous Polymeric Composites

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An analytical approach on the evaluation of interphase Poisson's ratio for fibrous composites, consisting of polymeric matrix and unidirectional continuous fibers, is performed. The simulation of the microstructure of the composite was carried out by means of a modified form of Hashin-Rosen cylinder assemblage model. Next, by the use of this three-phase model the authors impose some limitations to the polynomial variation laws which are commonly adopted to approximate the thermomechanical properties of the interphase layer of this type of polymeric composites and then propose an nth-degree polynomial function to approximate the Poisson's ratio of this layer.

1. Introduction

The rigorous description of a composite system consisting of a matrix in which continuous or short fibers have been dispersed is not an easy task. Indeed, a great number of parameters, geometrical, topological, mechanical, etc. are necessary, the majority of which vary in a stochastic manner or are practically unknown. Theoretical treatments usually attempt to exploit as far as possible readily available information, which, in most cases, consists of the mechanical properties of matrix and fiber and the volume fraction of the latter, whilst appropriate assumptions cover missing data. On the other hand it is well known that in lightweight structures, large specific stiffness, and strength are aiming properties for a material. By combining fibers with an appropriate polymer and controlling the production procedure it is possible to manufacture composites featuring large specific properties. Moreover, the constituents are usually cheap and easily processed, e.g., by injection molding. Hence, fibrous composites have many industrial applications where high performance per weight at a reasonable price is required. One of the most useful forms of composites for the construction

of high-performance structural elements is the type of panels made from aligned fibers containing polymerized matrix. Evidently, their mechanical properties depend on the related properties and volume fractions of the constituent materials, the fiber length or aspect ratio, the degree of the alignment, the adhesion between fibers, and matrix and last but not least they are affected by the fabrication techniques [1]. As it was initially stated, the investigation of the elastic properties of unidirectional fibrous composites reinforced with long or short fibers in terms of the related properties of constituents constitutes a very difficult problem of applied mechanics. In a fundamental investigation by Hashin and Rosen [2] bounds and explicit expressions to estimate the effective elastic moduli of unidirectional fibrous composites were derived via a rigorous variational method. On the other hand, amongst the analytical models presented in the literature, some of them take into consideration the existence of an intermediate phase developed during the preparation of the polymeric composite. Evidently this new phase plays an important role in the overall thermomechanical behavior of the composite.

In the meanwhile, the existence of a boundary interphase in polymeric composites (fibrous and particulate) was shown

experimentally by Lipatov [3] who measured its thickness by means of Differential Scanning Calorimetry (DSC) experiments. Also, in this valuable investigation, it was empirically stated that the size of these heat capacity jumps for unfilled and filled polymers is expressed in terms of the thickness of this phase. Yet, the estimation of its physical properties was a remaining problem.

To this end, in a preliminary model performed by Papanicolaou et al. [4] and Theocaris et al. [5] this intermediate phase was initially assumed to be a homogeneous and isotropic material whereas in a better approximation [6] a more complex model was introduced, according to which the fiber was surrounded by a series of successive cylinders, each one of them having a different elastic modulus in a step-function variation with respect to the polar radius.

In addition, problems of obtaining closed-form solutions to many cases of elastic inclusions embedded in an elastic matrix were encountered by Muskhelishvili [7] by the use of conformal mapping techniques. In addition, numerical solution techniques such as finite difference and finite elements have been extensively used to this end [8, 9]. Another remarkable consideration of the variable modulus interphase is the so-called unfolding model, which was carried out on the basis that the boundary interphase layer constitutes a transition zone between fibers with high moduli and matrix with rather low stiffness [10]. An extension of the above fundamental model to more complex composite systems (fibrous and particulate) was exhibited by Theocaris in [11].

In addition, a remarkable study performed by Swain et al [12] has thrown light on the effect of the boundary interphase on the properties and performance of composite materials and laminates.

In the past years, there is a lot of recent research work carried out for the evaluation of elastic and thermal properties of fibrous composites and for the investigation of the influence of various parameters on these properties. A detailed study on the effect of interphase layers on local thermal displacements in fibrous composites was presented in [13]. Also, in [14], an investigation was made by the aid of strength of materials and elasticity approach, to derive explicit expressions for longitudinal, transverse, and shear moduli along with longitudinal and transverse Poisson's ratio using the concept of boundary interphase between fiber and matrix, in the framework of a three-phase modified form of Hashin-Rosen cylinder assemblage model that was presented in [2]. Here, to evaluate the elastic properties of interphase a unified mode of variation was considered. In particular, an nth degree polynomial variation was initially adopted in terms of the radius of the introduced three-phase coaxial model and finally for facility reasons a quadratic function was taken into account. The above approach of interphase unknown properties was continued and extended to estimate the thermal conductivity of the same class of fibrous composites in [15]. In this work, to cover the whole spectrum of interphase thermal conductivity, five different laws of variation were used of which only two constitute polynomial forms (linear and parabolic). Nevertheless, it can be said that a three-layer cylindrical model to simulate

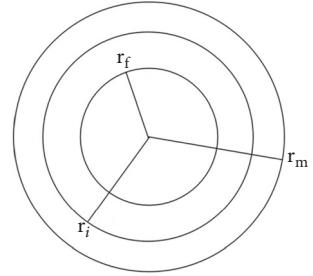


FIGURE 1: Coaxial three-phase cylinder unit cell.

the structure of unidirectional fibrous composites reinforced with continuous fibers may arise based on the theory of self-consistent models and adapting this approach to a three-layered cylinder, delineating the representative volume element for the fibrous composite [11]. Here one should elucidate that a significant variation of the self-consistent model is the three-phase model first introduced by Kerner [16], according to which the inclusion is developed by a matrix annulus which in turn is embedded in an infinite medium with the unknown macroscopic properties of the composite. Further, in [17, 18] the adoption of linear variation laws for interphase elastic constants yielded theoretical predictions of the composite moduli in a reasonable accordance with experimental values. Yet, to approach the interphase modulus and Poisson's ratio by a quadratic (parabolic) law with respect to the radius of the adopted model of embedded cylinders is many times more convenient since many times the linear variation law cannot alleviate the fact that the transition of the elastic constants from the matrix to fiber is carried out by "jumps" in their characteristic properties [17, 19]. However, a significant problem that remains is the following: letting an interphase property $M_i(r)$ be approached by a quadratic polynomial in the general form $Ar^2 + Br + C$, there are three coefficients $A; B; C$ needed to be found. Unfortunately, in the context of the modified form of Hashin-Rosen model introduced in [14] there are only two boundary conditions available. Specifically, at $r = r_f : M_i(r) = M_f$ and at $r = r_i : M_i(r) = M_m$. Another condition may arise by requiring the first derivative $dM_i(r)/dr$ to vanish at a critical point over the interval $[r_f, r_i]$ which corresponds to a given local extremum (maximum or minimum) [14]. In particular it was assumed that the critical point coincides with one of the endpoints of this interval and thus $dM_i(r_f)/dr = 0 \vee dM_i(r_i)/dr = 0$. Nevertheless, should this critical point be outside this interval or in the interval (r_f, r_i) such a condition cannot hold [20] and therefore the coefficients $A; B; C$ are unable to be calculated. To overcome this unfortunate situation, in the present work an alternative approach on the estimation of interphase elastic constants is proposed.

2. Analysis

Let us simulate the microstructure of a unidirectional fibrous composite by means of a coaxial three-phase cylinder unit cell, the cross-sectional area of which can be seen in Figure 1.

Here the radii $r_f; r_i; r_m$ denote the bounds of each separate zone, i.e., fiber, interphase, and matrix, respectively. The fiber zone begins at the zero value of the radius of the three-phase cylindrical model and ends at $r = r_f$. Next, the interphase zone, starts marginally at $r \geq r_f$ and ends at $r = r_i$. Finally, the matrix zone starts marginally at $r \geq r_i$ and finishes at $r = r_m$.

Evidently, the domain of definition of any single-valued continuous function that can be selected to approach the Poisson's ratio of this intermediate phase between fiber and matrix is the interval $[r_f, r_i]$.

The above modified form of Hashin-Rosen cylinder assemblage model was initially used in [14]. In the sense of the above-mentioned work the elastic constants of the interphase, i.e., stiffness, shear modulus, and Poisson's ratio can be generally approached as nth degree polynomials with respect to the polar radius r of the above coaxial three-phased model of embedded cylinders.

Suggestively, let us focus on Poisson's ratio of the interphase zone and express it with respect to radius r of the above model, as follows:

$$\nu_i(r) = Ar^n + Br^{n-1} + Cr^{n-2} + \dots \quad (1)$$

$$\text{with } r_f \leq r \leq r_i \quad (1a)$$

$$\text{and } \nu_f \leq \nu_i(r) \leq \nu_m \quad (1b)$$

Hence, (1) denotes the mode of variation for Poisson's ratio of this intermediate phase the thickness of which is evidently $r_i - r_f$ and can be experimentally measured as we shall mention later on.

For facility reasons and to be in accordance with inequality one may consider a polymeric fibrous composite the constituents of which are epoxy resin and e-glass fibers, i.e., $\nu_f = 0.2$; $\nu_m = 0.35$.

Moreover, at the interfaces amongst the three different phases of the overall material the following boundary conditions hold [14]:

$$\text{At } r = r_f: \quad \nu_i(r) = \nu_f \quad (2)$$

$$\text{At } r = r_i: \quad \nu_i(r) = \nu_m \quad (3)$$

Here on the basis of inequalities and one may pinpoint that should a linear variation law be adopted to approach the Poisson's ratio of this layer; the single valued function of the form $\nu_i(r) = Ar + B$ is strictly increasing. Yet, as stated above such an approach constitutes an oversimplified mode of variation.

In continuing, let us consider the following general quadratic function to approximate the interphase Poisson's ratio:

$$\nu_i(r) = Ar^2 + Br + C \quad (4)$$

with $A; B; C$ being arbitrarily selected real constants

Now, the mode of variation for the Poisson's ratio of the boundary interphase is reduced to a second-degree polynomial function in terms of polar radius r of the cylindrical three-phase model.

Nonetheless, the remaining problem is the determination of the three coefficients $A; B; C$ given that only two boundary conditions are generally available, i.e., (2) and (3).

Now, without violating the generality, let us set

$$\begin{aligned} A &\longrightarrow \frac{a}{4} \\ B &\longrightarrow a \cdot b \\ C &\longrightarrow a \cdot b^2 + c \end{aligned} \quad (5)$$

where a, b, c are arbitrary real numbers

Thus (4) becomes

$$\nu_i(r) = \frac{a}{4}r^2 + a \cdot br + (a \cdot b^2 + c) \quad (6)$$

Here one may observe that the latter relationship is completely synonymous to (4) because the totally arbitrary selection of the constant quantities a, b, c from the set of real numbers prohibits any correlation amongst the initial coefficients A, B, C appearing in (4).

Next, by differentiating the above relation with respect to polar radius r , one finds

$$\frac{d}{dr}\nu_i(r) = a\left(\frac{r}{2} + b\right) \quad (7)$$

and therefore

$$\begin{aligned} \frac{1}{a} \cdot \frac{d}{dr}\nu_i(r) &= \frac{r}{2} + b \iff \\ \frac{1}{a^2} \cdot \left(\frac{d}{dr}\nu_i(r)\right)^2 &= \left(\frac{r}{2} + b\right)^2 \iff \\ \frac{1}{a^2} \cdot \left(\frac{d}{dr}\nu_i(r)\right)^2 &= \frac{r^2}{4} + rb + b^2 \iff \\ \frac{1}{a} \cdot \left(\frac{d}{dr}\nu_i(r)\right)^2 &= \frac{a}{4}r^2 + abr + rb^2 \end{aligned} \quad (8)$$

Equation (8) can be combined with (6) to yield

$$\begin{aligned} \frac{1}{a} \cdot \left(\frac{d}{dr}\nu_i(r)\right)^2 &= \nu_i(r) - c \iff \\ \left(\frac{d}{dr}\nu_i(r)\right)^2 &= a \cdot (\nu_i(r) - c) \end{aligned} \quad (9)$$

Evidently, (9) constitutes a first-order separable ordinary differential equation of the form $N(y)y' = Q(x)$ or $N(y)dy = Q(x)dx$ and can be solved by trivial techniques [21].

Also, one observes that should the term in the left-hand side of (9) be nonzero the terms $a; (\nu_i(r) - c)$ agree in sign.

Now, to find the critical value of the variable r where the single-valued continuous function $\nu_i(r)$ has local extremum

(minimum or maximum) the quantity $(d/dr)\nu_i(r)$ should be set equal to zero. Thus it implies that

$$\begin{aligned} \left(\frac{d}{dr}\nu_i(r) \right) = 0 &\iff \\ a\left(\frac{r}{2} + b\right) = 0 &\iff \\ r_{cr} = -2b \end{aligned} \quad (10)$$

Besides, at this critical point, (9) yields

$$\begin{aligned} \left(\frac{d}{dr}\nu_i(r_{cr}) \right)^2 = 0 &\iff \\ a \cdot (\nu_i(r_{cr}) - c) = 0 &\iff \\ \nu_i(r_{cr}) = c \end{aligned} \quad (11)$$

Next, the application of the boundary conditions, expressed by (2) and (3), to (9) in association with (11) yields

$$\left(\frac{d}{dr}\nu_i(r_f) \right)^2 = a \cdot (\nu_f - \nu_i(r_{cr})) \quad (12)$$

and

$$\left(\frac{d}{dr}\nu_i(r_m) \right)^2 = a \cdot (\nu_m - \nu_i(r_{cr})) \quad (13)$$

Here, one may conclude that should the terms in the left-hand sides of (12) and (13) be strictly positive the terms $a \cdot (\nu_f - \nu_i(r_{cr}))$ and $a \cdot (\nu_m - \nu_i(r_{cr}))$ agree in sign.

In this framework, one infers that should the derivatives $(d/dr)\nu_i(r_f)$ and $(d/dr)\nu_i(r_m)$ be non-zero, the terms $(\nu_f - \nu_i(r_{cr}))$; $(\nu_m - \nu_i(r_{cr}))$ agree in sign.

In other words, since $r_{cr} = -2b$, if $r_f \neq -2b$ and $r_m \neq -2b$, the local extremum $\nu_i(r_{cr})$ does not belong to the interval $[\nu_f, \nu_m]$.

However, given a polymeric composite consisting of matrix and filler (particles or fibers), the values of interphase properties can be neither greater nor less than those of the constituents' properties [4, 11].

Thus if the critical point r_{cr} yields a value of interphase Poisson's ratio outside the interval $[\nu_f, \nu_m]$ the three coefficients $A; B; C$ appearing in (4) or $a; b; c$ in (6) should be calculated only on the basis of the boundary conditions designated by (2), (3) and the validity of any condition concerning the vanishing of $((d/dr)\nu_i(r))$ over the interval $[r_f, r_i]$ cannot be required.

In addition, by focusing on (4) and taking into account the basic concepts of Polynomial theory [22], one may obtain the critical point of $\nu_i(r)$ by the following relation which holds identically:

$$\nu_i\left(-\frac{B}{2A}\right) \equiv \frac{4AC - B^2}{4A} \quad (14)$$

Evidently the fraction on the right hand side of the above identity signifies the unique local extremum of the quadratic

variation law of Poisson's ratio. Also one may point out that (14) is synonymous to the vanishing of the first derivative of ν_i since the determination of the critical point can be carried out by setting $(d/dr)\nu_i(r) = 0$.

In this context, in order to specify a third condition to complete (2),(3) towards the calculation of the three coefficients A, B, C or a, b, c one should know beforehand either the value of r_{cr} in terms of the endpoints r_f, r_i or the value of the local extremum in terms of the values corresponding at the endpoints, i.e., ν_f, ν_m with $\nu_f < \nu_m$. In [14] it was stated that should the value $(4AC - B^2)/4A$ denote a local minimum it coincides with ν_f whereas should denote a local maximum it coincides with ν_m . Yet it is the authors' current opinion on this issue that this assumption is oversimplified and rather unrealistic, although it has yielded theoretical predictions for the composite elastic properties that were found to be in good agreement with several reliable theoretical formulae and experimental data.

Hence should one select a second degree parabola to approach the interphase Poisson's ratio expressed either by (4) or (6), except the two boundary conditions concerning the interface with fiber and matrix, respectively, another condition is necessary referring either to the critical point of the quadratic function or the value of interphase Poisson's ratio yielded by this critical point. Evidently, the fact that (14) holds identically prohibits the parallel use of the two above additional conditions designated by (10) and (11), which in general can be presented by the following two expressions:

$$r_{cr} = r_f * r_i \quad (15)$$

Here, the notation $*$ denotes a binary operation over the set of real numbers combining the elements r_f, r_i to produce univocally another real number.

$$\nu_i(r_{cr}) = \nu_f \bullet \nu_m \quad (16)$$

Similarly, the notation \bullet designates a binary operation over the set of real numbers combining the elements ν_f, ν_m to produce univocally another real number.

Moreover since $r_f < r_i$ and $\nu_f < \nu_m$ these binary operations should not be necessarily commutative.

In this context, should the critical value r_{cr} yielded by the operation $r_f * r_i$ lie on the interval $[r_f, r_i]$ it is necessary to require the corresponding value of Poisson's ratio $\nu_i(r_{cr}) = \nu_i(r_f * r_i)$ to be strictly greater than ν_f and strictly less than ν_m .

On the other hand, should the value of Poisson's ratio $\nu_i(r_{cr}) = \nu_f \bullet \nu_m$ be strictly less than ν_f or strictly greater than ν_m it is necessary to require the value r_{cr} to be outside the interval $[r_f, r_i]$.

Obviously, similar analyses on the basis of the same reasoning could take place for the other elastic or thermal properties of interphase region, i.e., stiffness, shear modulus, thermal expansion coefficient etc.

Next, in the sense of [14] let us introduce a general third-degree polynomial to approximate the interphase Poisson's ratio given as

$$\nu_i(r) = Ar^3 + Br^2 + Cr + D \quad (17)$$

Evidently, (17) signifies a more complicated mode of variation for interphase Poisson's ratio when compared with (4).

Yet, it is known from Calculus [20] that if $B^2 - 3AC \leq 0$ this function will have no local extrema over its domain of definition and therefore, except (2) and (3), no additional conditions can be set regarding the first derivative of $\nu_i(r)$.

Hence, the consideration of a general third-degree polynomial to approach the interphase Poisson's ratio or any other elastic property of this phase can be made on the premise that the quantity $B^2 - 3AC$ is strictly positive.

Thus we can write out

$$\frac{d}{dr} \nu_i(r) = 3Ar^2 + 2Br + C \quad (18)$$

Provided that $B^2 - 3AC > 0$ the critical points are given as

$$r_{cr,1} = \frac{-Br - \sqrt{B^2 - 3AC}}{A}; \quad (19a)$$

$$r_{cr,2} = \frac{-Br + \sqrt{B^2 - 3AC}}{A} \quad (19b)$$

Thus in a quite analogous reasoning with that resulting in (15), (16) one may require the validity of the following additional conditions:

$$r_{cr,1} = r_f * r_i; \quad (20a)$$

$$r_{cr,2} = r_f \diamond r_i \quad (20b)$$

or

$$\nu_i(r_{cr,1}) = \nu_f \bullet \nu_m; \quad (21a)$$

$$\nu_i(r_{cr,2}) = \nu_f \circ \nu_m \quad (21b)$$

Apparently the above disjunction is exclusive. Now, should even one of the critical values $r_{cr,1}$; $r_{cr,2}$ yielded by the operations $r_f * r_i$ and $r_{cr,2} = r_f \diamond r_i$, respectively, lie on the interval $[r_f, r_i]$ it is necessary to require the corresponding value of Poisson's ratio to be strictly greater than ν_f and concurrently strictly less than ν_m .

On the other hand, should even one the values of Poisson's ratio $\nu_i(r_{cr,1}) = \nu_f \bullet \nu_m$; $\nu_i(r_{cr,2}) = \nu_f \circ \nu_m$ be strictly less than ν_f or strictly greater than ν_m it is necessary to require the corresponding critical point to lie outside the interval $[r_f, r_i]$.

Moreover, on the basis of (17), it can be proved that the following relationship holds:

$$\begin{aligned} \nu_i(r_{cr,1}) \cdot \nu_i(r_{cr,2}) &= \left(\frac{2B^3}{27A^2} - \frac{BC}{3A} + D \right)^2 \\ &\quad + \frac{16}{27A} \left(C - \frac{B^2}{3A} \right)^3 \end{aligned} \quad (21c)$$

For a thorough presentation of the mathematical derivations resulting in (21c) let us refer to Appendix Section.

Nevertheless, since (15), (16), (20a), (20b), (21a), (21b), (21c) have a rather theoretical character let us give a more specific example of the necessity of these conditions by

proposing the following single-valued polynomial representation to approximate interphase Poisson's ratio

$$\nu_i(r) = A(\nu_m - \nu_f)(r - r_f)^n (r_i - r)^n + Br + C \quad (22)$$

with $n \in N^*$ and A, B, C being arbitrary real numbers

An application of the boundary conditions expressed by (2) and (3) at the interfaces with fiber and matrix, respectively, yields

$$Br_f + C = \nu_i(r_f) = \nu_f \quad (23)$$

$$Br_i + C = \nu_i(r_i) = \nu_m \quad (24)$$

and therefore

$$B = \frac{\nu_m - \nu_f}{r_i - r_f} \quad (25)$$

$$C = \nu_f - \frac{\nu_m - \nu_f}{r_i - r_f} r_f \quad (26)$$

In the sequel, let us calculate the first derivative of $\nu_i(r)$ with respect to polar radius

$$\begin{aligned} \frac{d\nu_i(r)}{dr} &= nA(\nu_m - \nu_f)(r - r_f)^{n-1}(r_i - r)^n \\ &\quad - nA(\nu_m - \nu_f)(r_i - r)^{n-1}(r - r_f)^n + B \end{aligned} \quad (27)$$

and therefore

$$\begin{aligned} \frac{1}{nA(\nu_m - \nu_f)} \left(\frac{d\nu_i(r)}{dr} - B \right) \\ &= (r - r_f)^{n-1}(r_i - r)^n - (r_i - r)^{n-1}(r - r_f)^n \iff \\ \frac{1}{nA(\nu_m - \nu_f)} \left(\frac{d\nu_i(r)}{dr} - B \right) \\ &= (r - r_f)^{n-1}(r_i - r)^{n-1}(r_f + r_i - 2r) \iff \\ \frac{1}{2nA(\nu_m - \nu_f)} \left(\frac{d\nu_i(r)}{dr} - B \right) \\ &= \frac{1}{2}(r_f + r_i - 2r)(r - r_f)^{n-1}(r_i - r)^{n-1} \iff \\ \frac{1}{2nA(\nu_m - \nu_f)} \left(\frac{d\nu_i(r)}{dr} - B \right) \\ &= (r - r_f)^{n-1}(r_i - r)^{n-1}(M_{Ar}(r_f, r_i) - r) \end{aligned} \quad (28)$$

Here $M_{Ar}(r_f, r_i)$ denotes the arithmetic mean of r_f, r_i .

Equation (28) can be combined with (25) to yield

$$\begin{aligned} \frac{1}{2nA(\nu_m - \nu_f)} \left(\frac{d\nu_i(r)}{dr} - \frac{\nu_m - \nu_f}{r_i - r_f} \right) \\ &= (r - r_f)^{n-1}(r_i - r)^{n-1}(M_{Ar}(r_f, r_i) - r) \end{aligned} \quad (29)$$

At $r = r_{cr}$ one obtains

$$\begin{aligned} \frac{1}{2nA(\nu_m - \nu_f)} \left(-\frac{\nu_m - \nu_f}{r_i - r_f} \right) \\ &= (r_{cr} - r_f)^{n-1}(r_i - r_{cr})^{n-1}(M_{Ar}(r_f, r_i) - r_{cr}) \end{aligned} \quad (30)$$

By focusing on the above representation one may point out that the critical points r_{cr} at which $d\nu_i(r)/dr$ vanishes cannot coincide with the endpoints r_f, r_i neither with their arithmetic mean.

On the other hand, in an analogous manner with that resulting in (9) let us try to relate in an explicit manner the interphase Poisson's ratio to its first derivative.

In this context, (27) yields

$$\begin{aligned} \frac{d\nu_i(r)}{dr} - B \\ = nA(\nu_m - \nu_f)(r - r_f)^{n-1}(r_i - r)^n \\ - nA(\nu_m - \nu_f)(r_i - r)^{n-1}(r - r_f)^n \iff \\ \frac{d\nu_i(r)}{dr} - B \\ = A(\nu_m - \nu_f)(r - r_f)^n(r_i - r)^n \left(\frac{n}{r - r_f} - \frac{n}{r_i - r} \right) \end{aligned} \quad (31)$$

Now, given that the coefficient B has been already calculated, the following relation links the critical points r_{cr} , where $d\nu_i(r)/dr$ vanishes, with the coefficient A

$$\begin{aligned} B = A(\nu_m - \nu_f)(r_{cr} - r_f)^n(r_i - r_{cr})^n \\ \cdot \left(\frac{n}{r_i - r_{cr}} - \frac{n}{r_{cr} - r_f} \right) \end{aligned} \quad (32)$$

Equation (32) can be combined with (22) to yield

$$B = (\nu_i(r_{cr}) - Br_{cr} - C) \cdot \left(\frac{n}{r_i - r_{cr}} - \frac{n}{r_{cr} - r_f} \right) \quad (33)$$

Solving for $\nu_i(r_{cr})$ one obtains

$$\nu_i(r_{cr}) = \frac{B}{(n/(r_i - r_{cr}) - n/(r_{cr} - r_f))} + Br_{cr} + C \quad (34)$$

Equation (34) can be combined with (25) and (26) to yield explicitly the set of the local extrema of interphase Poisson's ratio in terms of the polynomial degree n . Thus we can write out

$$\begin{aligned} \nu_i(r_{cr}) &= \frac{(\nu_m - \nu_f)/(r_i - r_f)}{(n/(r_i - r_{cr}) - n/(r_{cr} - r_f))} + \frac{\nu_m - \nu_f}{r_i - r_f}r_{cr} \\ &+ \nu_f - \frac{\nu_m - \nu_f}{r_i - r_f}r_f \iff \\ \nu_i(r_{cr}) &= \frac{(\nu_m - \nu_f)/(r_i - r_f)}{(n/(r_i - r_{cr}) - n/(r_{cr} - r_f))} \\ &+ \nu_f \frac{\nu_m - \nu_f}{r_i - r_f}(r_{cr} - r_f) \end{aligned} \quad (35)$$

Here one observes that the set of local extrema of interphase Poisson's ratio has been evaluated without the knowledge of the coefficient A which ought to be calculated by (30) or by (32).

Evidently, these two equations are equivalent and cannot be considered as a system.

Right here is the necessity of the theoretical representations designated by (15), (16), (20a), (20b) and (21a), (21b), (21c).

In [14] it was assumed in a rather subjective and very simplified mode that $\nu_i(r_{cr}) = \nu_f \vee \nu_i(r_{cr}) = \nu_m$ whilst $r_{cr} = r_i \vee r_{cr} = r_f$.

Apparently, such a consideration is rather unrealistic. However, a reasonable conjecture for the calculation of coefficient A is to focus on (30) or (35) which yields the Poisson's ratio in a closed form and try to search for the critical points $r = r_{cr}$ by means of other types of means as discussed in [23] since it has been proved that mean-theory is very useful both from theoretical point of view and for practical (engineering) purposes. Yet, such an approach cannot include the case when $r_{cr} < r_f$ or $r_{cr} > r_i$ and then one may examine the interval $[0, r_m]$. Alternatively, given that most materials (except those ones with auxetic core) have Poisson's ratio values ranging between 0 and 0.5, one may carry out a parametrical study considering an upper bound of interphase Poisson's and then substitute it on (35) in order to solve it for r_{cr} .

3. Discussion

The boundary interphase zone was assumed as a natural phase which is developed in reality between fiber and polymer matrix. In this context, it can be said that this intermediate phase is neither an artificial one, e.g., by the immersion of the fibers in an agent, nor a pseudophase being contrived to simulate the microstructure of the composite. Hence it is not possible to know beforehand or to determine the interphase properties, a fact that renders necessary to make assumptions about them. In order to approximate the mode of variation of the variable elastic properties of interphase layer, such as Poisson's ratio, an n th degree polynomial function was considered by one of the authors in [14]. This function for $n = 2$ yielded a parabolic law. Also, to assume that at $r = r_i$, i.e. at the interface between matrix and interphase, $\nu_i(r_i) = \nu_m$ is realistic indeed. Moreover, the assumption that at $r = r_f$, i.e., at the interface between fiber and interphase $\nu_i(r_f) = \nu_f$ seems reasonable too. However, the remaining problem is to find the coefficients of such a polynomial function given that only the above two boundary conditions are available. The vanishing of the first derivative of interphase Poisson's ratio (or stiffness, shear modulus, etc.) at the endpoints of the interval $[r_f, r_i]$ that was adopted in [14] seems to be a rather superficial and oversimplified consideration though it yielded realistic theoretical predictions for the elastic constants of the composite.

In the current investigation the authors made an endeavor to shed some light on this difficult task by setting some limitations to the polynomial variation laws of second and third degree in the form of some theoretical formulae expressed by (15), (16), (20a), (20b), (21a), (21b), (21c) and in the sequel they propose an n th degree polynomial function to approximate the variation of interphase Poisson's ratio.

In this framework, this fundamental elastic property can be estimated provided of course an experimental estimation of the interphase thickness $\Delta r = r_i - r_f$ and given that r_f is known beforehand.

To illustrate the physical meaning of Δr , one may emphasize that in reality any polymeric composite (fibrous or particulate) necessarily consists of three distinct phases (matrix, filler, and interphase) as it is stated in [3, 11].

Lipatov [3] 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. Apparently, as the filler volume fraction is increased, the proportion of macromolecules characterized by a reduced mobility is also increased. This is equivalent to an augmentation of the interphase concentration by volume and evidently it is in consensus with the conclusion of [3] that the extent of interphase expressed by its thickness Δr is the reason of the variation of the amplitudes of heat capacity jumps appearing at the glass transition zones of the matrix material and the composite with various fiber contents. The size of heat capacity jumps for unfilled and filled materials is directly related to Δr by the next empirical relationship:

$$\frac{(r_f + \Delta r)^2}{r_f^2} - 1 = \frac{\lambda U_f}{1 - U_f} \quad (36)$$

where the coefficient λ is evaluated as

$$\lambda = 1 - \frac{\Delta C_p^f}{\Delta C_p^0} \quad (37)$$

Here the numerator and the denominator of the fraction in the right-hand side of (37) are the sudden changes of the heat capacity for the reinforced polymer and pure one, respectively.

In this context, by using Lipatov's theory [3] interrelating the abrupt jumps in the specific heat of a composite at its respective glass transition temperatures with the values of the extent of this boundary layer, the interphase thickness can be calculated in an accurate manner and thus the proposed analytical method for the theoretical determination of Poisson's ratio of this unknown intermediate phase is further enhanced.

4. Conclusions

This paper, as a continuation into a previous research work carried out by one of the authors, had two main objectives:

The first aim was to set some restrictions to the use of a single-valued polynomial function of nth degree to approach the interphase elastic properties for a general class of fibrous composites.

These restrictions concern the set of critical points and the corresponding local extrema (minima and /or peak points).

A modified form of the well-known Hashin-Rosen cylinder assemblage model was adopted to simulate the microstructure of the overall material.

Suggestively, the authors focused on interphase Poisson's ratio and examined in a rigorous theoretical manner its variations over the interval defined by the interphase thickness Δr , as obtained from a second and third degree parabola.

Next, the authors introduced an nth degree polynomial function with respect to the radius of the coaxial three-phase cylinder model in order to approach the interphase Poisson's ratio. This function renders very clear the necessity of the indicated restrictions for the set of critical points.

In this context, this fundamental elastic property of the interphase layer can be accurately estimated on the premise that an experimental measurement of the interphase thickness is carried out.

Appendix

In this section, let us perform in detail the mathematical derivations resulting in (21c) which links the maximum and minimum value of interphase Poisson's ratio when the latter is approached by a third-degree polynomial with respect to the radius of the coaxial three-phase cylindrical model that was adopted to simulate the composite structure.

Evidently, (17) can be formulated as

$$\nu_i(r) = A \left(r^3 + \frac{B}{A} r^2 + \frac{C}{A} r + \frac{D}{A} \right) \quad (A.1)$$

or equivalently

$$\nu_i(r) = A \left(r^3 + B_1 r^2 + C_1 r + D_1 \right) \quad (A.2)$$

Here, for facility reasons we have put

$$\begin{aligned} B_1 &= \frac{B}{A}; \\ C_1 &= \frac{C}{A}; \\ D_1 &= \frac{D}{A} \end{aligned} \quad (A.3)$$

Now, by the use of the trivial substitution $r = y - B_1/3$ it implies that

$$\begin{aligned} \nu_i(r) \equiv h(y) &= A \left(\left(y - \frac{B_1}{3} \right)^3 + B_1 \left(y - \frac{B_1}{3} \right)^2 \right. \\ &\quad \left. + C_1 \left(y - \frac{B_1}{3} \right) + D_1 \right) \end{aligned} \quad (A.4)$$

where $h : R \rightarrow R$ is a single-valued continuous function.

In addition, note that $y \in [r_f - B_1/3, r_i - B_1/3]$.

Thus we can write out

$$\begin{aligned} \frac{h(y)}{A} &= y^3 - 3y^2 \frac{B_1}{3} + 3y \frac{B_1^2}{9} - \frac{B_1^3}{27} \\ &\quad + B_1 \left(y - \frac{B_1}{3} \right)^2 + C_1 \left(y - \frac{B_1}{3} \right) + D_1 \iff \\ \frac{h(y)}{A} &= y^3 - y^2 B_1 + y \frac{B_1^2}{3} - \frac{B_1^3}{27} + B_1 y^2 - 2y \frac{B_1^2}{3} \\ &\quad + \frac{B_1^3}{9} + C_1 \left(y - \frac{B_1}{3} \right) + D_1 \iff \end{aligned} \quad (\text{A.5})$$

$$\begin{aligned} \frac{h(y)}{A} &= y^3 - \frac{B_1^3}{27} - y \frac{B_1^2}{3} + \frac{B_1^3}{9} + yC_1 - \frac{B_1}{3} C_1 \\ &\quad + D_1 \iff \\ \frac{h(y)}{A} &= y^3 + y \left(C_1 - \frac{B_1^2}{3} \right) + \frac{2B_1^3}{27} - \frac{B_1}{3} C_1 + D_1 \end{aligned}$$

and therefore

$$\begin{aligned} h(y) &= Ay^3 + y \left(AC_1 - \frac{AB_1^2}{3} \right) + \frac{2AB_1^3}{27} \\ &\quad - \frac{AB_1}{3} C_1 + AD_1 \end{aligned} \quad (\text{A.6})$$

Equation (A.6) can be combined with (A.3) to yield

$$h(y) = Ay^3 + \left(C - \frac{B^2}{3A} \right) y + \frac{2B^3}{27A^2} - \frac{BC}{3A} + D \quad (\text{A.7})$$

Now, for facility reasons, let us set

$$\begin{aligned} a &= A; \\ p &= C - \frac{B^2}{3A}; \\ q &= \frac{2B^3}{27A^2} - \frac{BC}{3A} + D \end{aligned} \quad (\text{A.8})$$

Thus one may deduce that

$$h(y) = ay^3 + py + q \quad (\text{A.9})$$

The first derivative with respect to auxiliary variable y is given as

$$\frac{dh(y)}{dy} = 3ay^2 + p \quad (\text{A.10})$$

Evidently, the necessary and sufficient condition in order for the quantity $dh(y)/dy$ to have real roots is the coefficients a and p to disagree in sign. In this framework, one also infers that these real roots should disagree in sign too. Also, since $d^2h(y)/dy^2 = 6ay$, it implies that the polynomial $h(y)$ has a unique local minimum and a unique local maximum.

Let $y_1; y_2$ be the real roots of (A.10):

$$\begin{aligned} \text{Evidently, } y_1 &= \sqrt{-\frac{p}{3a}}; \\ y_2 &= -\sqrt{-\frac{p}{3a}} \end{aligned} \quad (\text{A.11})$$

Then without violating the generality we can write out

$$\max h(y) = \frac{p}{3} \sqrt{-\frac{p}{3a}} + p \sqrt{-\frac{p}{3a}} + q \quad (\text{A.12})$$

and

$$\min h(y) = -\frac{p}{3} \sqrt{-\frac{p}{3a}} - p \sqrt{-\frac{p}{3a}} + q \quad (\text{A.13})$$

and therefore

$$\begin{aligned} \max h(y) \cdot \min h(y) &= - \left(\frac{p}{3} \sqrt{-\frac{p}{3a}} + p \sqrt{-\frac{p}{3a}} + q \right) \\ &\quad \cdot \left(\frac{p}{3} \sqrt{-\frac{p}{3a}} + p \sqrt{-\frac{p}{3a}} - q \right) \iff \\ \max h(y) \cdot \min h(y) &= - \left(\left(\frac{p}{3} \sqrt{-\frac{p}{3a}} + p \sqrt{-\frac{p}{3a}} \right)^2 - q^2 \right) \iff \\ \max h(y) \cdot \min h(y) &= q^2 + \frac{16}{27} \frac{p^3}{a} \end{aligned} \quad (\text{A.14})$$

In continuing, the above relation can be combined with (A.8) to yield

$$\begin{aligned} \max h(y) \cdot \min h(y) &= \left(\frac{2B^3}{27A^2} - \frac{BC}{3A} + D \right)^2 + \frac{16}{27A} \left(C - \frac{B^2}{3A} \right)^3 \end{aligned} \quad (\text{A.15})$$

Finally, since $\gamma_i(r) \equiv h(y)$ one may deduce that

$$\begin{aligned} \gamma_i(r_{cr,1}) \cdot \gamma_i(r_{cr,2}) &= \left(\frac{2B^3}{27A^2} - \frac{BC}{3A} + D \right)^2 \\ &\quad + \frac{16}{27A} \left(C - \frac{B^2}{3A} \right)^3 \end{aligned} \quad (\text{A.16})$$

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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