A four-layered self-consistent scheme applied to two-phase composites

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A FOUR-LAYERED SELF-CONSISTENT SCHEME APPLIED TO TWO-PHASE COMPOSITES

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Introduction

For both particulate and unidirectional composites, the traditional scheme describing one phase as the matrix and the other as the reinforcement is not completely satisfactory for high reinforcement content. This article proposes the use of De Larrard and Le Roy's geometric model [1] to better define the morphological characteristics of a two-phase composite, in conjunction with the self-consistent method, thanks to the recent development of n+1 layered models. The comparison of results with experimental data has proved promising.

Analysis

The evaluation of the elastic properties of biphased materials uses homogenization techniques based on geometrical, physical and mechanical parameters. The prediction of the viscoelastic behavior of polymer matrix composites is based on these same models via Lee-Mandel's correspondence principle [2]. Among these approaches, several propose a particular space distribution of components, for instance the periodical homogenization methods. Self-consistent approaches are linked with a perfect random order situation [3]. Christensen and Lo [4] solved the problem of determining the transverse shear modulus for biphased composites; more recently, this technique was extended to n-phase composites by Hervé and Zaoui [5] for particulate composites, using a recurrent method. These authors [6], along with Agbossou [7] for the transverse shear modulus, extended their technique to unidirectional composites, and all used the same recurrent method. For their part, Pastor and Nguyen Viet proposed a direct semi-analytical solution [8], leading to three linear systems and a bilinear system. We will apply this technique, quite simple to

use on any personal computer, to the unidirectional case, using Hervé and Zaoui's method for particulate composites.

Nevertheless, if these models produce satisfactory results for low reinforcement contents, one usually notices a considerable discrepancy between theoretical and experimental results for higher contents. For these contents, De Larrard and Le Roy [1] proposed an inventive geometrical model with an improved consideration of actual phase configuration, nevertheless followed by application of an approximate homogenization method. This article presents the elasticity and viscoelasticity results obtained by coupling n-phase models (Hervé and Zaoui [5], Pastor et al. [8]), and the geometrical distribution proposed by De Larrard and Le Roy [1]. An initial application of this work was presented by Saffré [9] for the case of biphased aluminium-tin alloy, with absolutely non-miscible components, where the retained homogenization model was an interphase model for particulate composites proposed by Maurer [10].

Geometrical model representation

Let us first consider a composite unit volume ($V_c = 1$). The particulate or unidirectional reinforcement content (V_r) of a composite is always limited by the dry reinforcement maximum compactness, called optimum compactness content ($V_{r\ max}$). This layout corresponds to the reinforcement and matrix inversion. For an increase in Vr, the probability is greater and greater that reinforcements will themselves aggregate, embedding a part of the matrix (V_{me}) (FIG. 1).

FIG. 1

Reinforcement aggregates in the matrix
(a) Real composite of volume V_c,
(b) Composite formed by reinforcements and embedded part of matrix of volume V

If V is the volume composed of both the reinforcement volume and the embedded matrix volume, then it is possible to write (1) :

$$V_{r\,max} = \frac{V_r}{V} \tag{1}$$

and for the embedded part of the matrix :

$$V_{m\,e} = (1 - V_{r\,max})\,V = (1 - V_{r\,max})\,\frac{V_r}{V_{r\,max}} \tag{2}$$

The rest of the matrix, which has a freer behaviour, has a volume expressed in (3).

$$V_{m\,l} = 1 - \frac{V_r}{V_{r\,max}} \tag{3}$$

One can then analyse the material as a new composite, whose matrix has the same nature as the initial one, whose volume is expressed in (3), and whose inclusion, an imaginary material composed of the embedded matrix in a reinforcement shell, has a volume equal to $V_r/V_{r\,max}$. This composite is represented by the new elementary cell in figure 2.

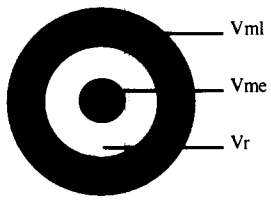

FIG. 2
Composite elementary cell.

De Larrard and Le Roy used this geometrical motif with Hashin's model [11], step by step, first solving the problem at the level of the two inner spheres, then adding the outer one.

Application

As we have pointed out in the analysis, recent extensions of the self-consistent scheme to n+1 phase models, make use of the above geometrical motif possible. Of course, these models can be brought back to four phases for which the representative elementary volume has the same features as the elementary cell represented in figure 2, which is in turn blurred into the infinite

homogenized medium. In the case of composites with particulate inclusions, bulk modulus is expressed in the following way :

$$k = k_m + \frac{(3k_m + 4\mu_m)(3k_m + 4\mu_r)}{\frac{(3k_r + 4\mu_r)(3k_m + 4\mu_m)}{V_r(k_r - k_m)} + 12\frac{V_{r\,max}}{V_r}(\mu_r - \mu_m) - 3(3k_m + 4\mu_r)} \quad (4)$$

where the m and r indexes respectively correspond to the matrix and the reinforcements.

Shear modulus is expressed as follows:

$$A\left(\frac{\mu}{\mu_m}\right)^2 + 2B\left(\frac{\mu}{\mu_m}\right) + C = 0 \quad (5)$$

with A, B and C only a function of V_r, $V_{r\,max}$, μ_r and μ_m.

For unidirectional reinforcement composites, the calculations are too complex to be described in this paper, and it should be referred to Pastor and Nguyen Viet [8] or Hervé and Zaoui [6].

Results and discussion

1 - Particulate composite in viscoelasticity

The material studied is a composite, whose matrix is a polystyrene resin, and whose reinforcements are glass beads with a diameter varying between 70 and 110 µm, with an unknown geometrical distribution. This leads us to determine the optimum compactness of beads experimentally. The optimum reinforcement content obtained in this way is about 64 % in volume. Complex moduli are calculated here using Lee-Mandel's correspondence principle [2], and the evaluation technique for the complex Poisson's ratio used by Nguyen Viet [12] on the basis of a proposal by Theocaris (Cf. [13]). The first evaluation of this ratio, unattainable experimentally, was proposed by Agbossou et al. [14]. These authors extended Theocaris's idea to complex moduli using a successive approximation technique. Nguyen Viet et al proved that this technique is equivalent to the bulk modulus being constant and real, i.e. that the matrix is only viscoelastic in shearing, as is often affirmed. The technique used here frees us from the latter approximation.

Figure 3 presents for comparison the values calculated with Hervé and Zaoui's technique [5] applied to the first defined geometrical model (curve a), the values calculated with Christensen and Lo's three-phase model [4] (curve b), as well as the experimental results (Fig. 3 - curve c), where E_m = 3.1 GPa, ν_m = 0.33, tan $\delta = 1.24.10^{-2}$, E_r = 70.0 GPa, ν_r = 0.2.

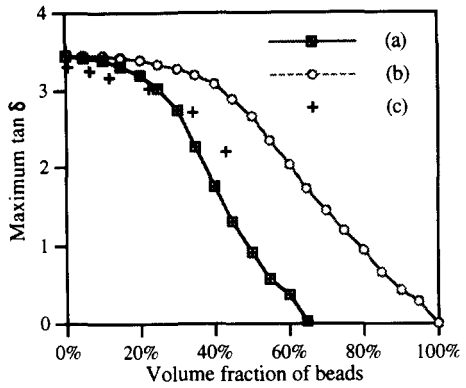

FIG. 3
Loss factor versus the volume fraction of beads

The results obtained, with high volume reinforcement contents, remain less than the experimental results. This is normal, since the calculation result may be considered as the lower boundary for the exact result, as the model implies that the reinforcements themselves aggregate, forming compact macro-inclusions, in a perfect random order situation, containing within themselves the practical minimum embedded matrix volume. Nevertheless, these results are more realistic than those produced by Christensen and Lo's model, if we consider that the composite loss factor is in actual fact negligible for bead contents greater than 64 %, a content which we earlier called the "practical optimum compactness content", where the blended beads limit the embedded matrix strain. The proposed method therefore takes into account as much as possible, in experimental fashion, the generally distributed aspect of the glass beads' granulometry.

2 - Unidirectional composite in elasticity

We also studied the case of a glass-epoxy composite with unidirectional reinforcements in transverse simple tension, for which experimental results were obtained by Lagache et al. [15]. Glass fibers have the same diameter, and the fibers' theoretical maximum compactness, which corresponds to a compact hexagonal stack, is equal to 90.7 %. This theoretical limit is never reached in practice, which is why we apply the user's well-known limit, also confirmed by Gutowski et al. [16], which is about 76 % in volume. In transverse isotropy, we chose Pastor and NGuyen's [8] homogenization model to solve our four-phase model. As for the case we studied first, Figure 4 presents the obtained results (curve a), compared with Christensen and Lo's model [4] (curve b), as well as with the experimental results (curve c), where E_m = 2.9 GPa, ν_m = 0.4, E_r = 73.0 GPa, ν_r = 0.22.

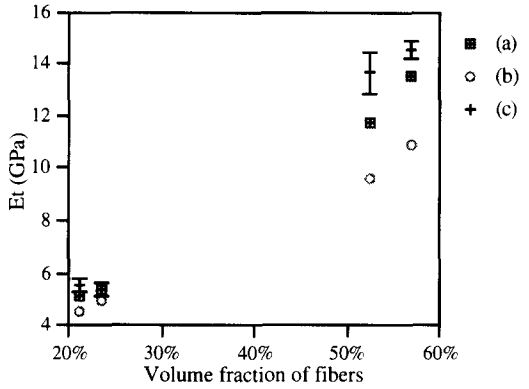

FIG. 4

Transverse Young modulus
versus the volume fraction of fibers.

Although we note a good match between the theoretical and experimental results for reinforcement contents of the order of 20 %, there remains a difference for higher reinforcement contents, but this average difference goes from 25 to 8 % when we use the model presented. The most realistic explanation for the remaining difference is a modification of the in situ matrix properties. Indeed, a simple increase in the matrix Young modulus from 2.9 to 3.2 GPa (a perfectly acceptable value for a strong reticulated epoxy network) brings theoretical values back within the range of the experimental results.

Conclusion

The main advantages of the model presented are its simplicity and the easy monitoring of the optimum compactness content. Furthermore, good quality results are obtained without any additional approximation, especially considering the simplicity of the model. Thus, we can affirm that the model presented considerably improves the ability to predict the mechanical characteristics of the biphased materials studied. Nevertheless, the approximation provided by four-phase models for high fibrous or particulate reinforcement contents still requires improvement.

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