Computer model for the elastic properties of short fibre and particulate filled polymers

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A finite difference type of approach is proposed for the study of the elastic properties of fibre and particulate reinforced composites. The effects of the fibre modulus E_f and aspect ratio (l/d) on the composite modulus are studied. In that respect, the ratio $(E_f/E_m)/(l/d)$, where E_m is the matrix modulus, is shown to be an important index of short fibre composites. Our results for the dependence of the composite modulus on E_f, l/d and volume fraction of the fibres are compared to available experimental data on foams and particulate filled polymers. A good agreement with experiment is found, which is much better than that obtained with the help of the widely used semi-empirical Halpin–Tsai equation.

1. Introduction

There has been recently a growing interest in fibre reinforced composites due to their great versatility and high performance. These materials often consist of discontinuous stiff fibres embedded in a soft matrix with the fibre axes oriented in the direction of the applied load. The physical and mechanical properties of these composites are, however, not well understood. The variation in fibre length as well as the interactions between the fibre ends and the matrix are the key factors contributing to the complexity of the problem. The present paper addresses a basic question: the effective moduli of these composites and their dependence on volume fraction, stiffness and aspect ratio of the fibres.

Several constitutive relationships or combining rules have been proposed for predicting the properties of reinforced composites in terms of the properties and concentrations of the constitutive components [1, 2]. These relationships lead to similar predictions for fibre volume fractions less than 20 to 30%. At higher volume fraction loadings, the results diverge and generally show poor agreement with experimental data because, e.g. of the neglect of fibre–fibre interactions. All these approaches, moreover, are only global in nature and are unable to provide a detailed representation of the stress concentration near fibre ends.

In a previous paper [3], we have developed a finite difference type of approach for the study of the stress transfer in fibre-reinforced composites. In that approach, the composite is represented by a regular three-dimensional lattice whose nearest neighbour nodal points are linked by bonds having different elastic constants for the fibre and for the matrix. For a given external strain, these nodes are relaxed towards local mechanical equilibrium with their

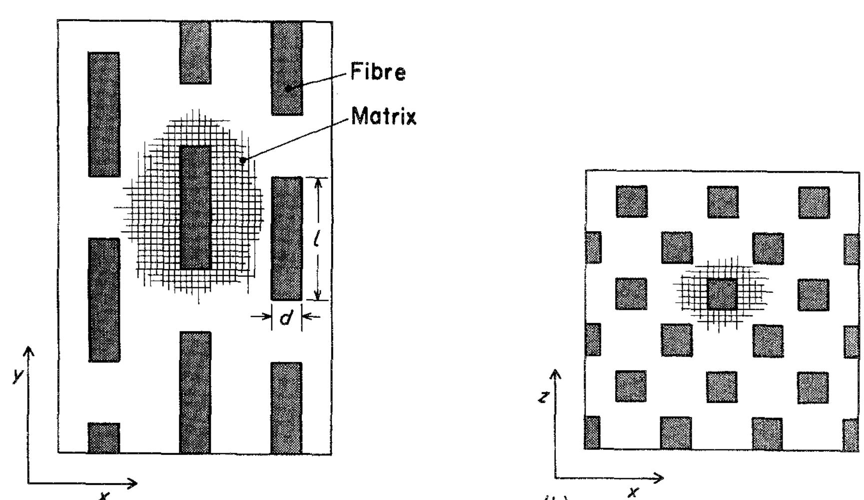

Figure 1 Two-dimensional representation of the lattice model in a plane parallel (Fig. 1a) and perpendicular (Fig. 1b), respectively, to the direction of loading (y-direction). The lattice comprises 120 nodes along the y-axis and 39 in the x- and z-directions. The parameters d and l denote the fibre diameter and length, respectively. In the simulations, d was set equal to 3 to 7 lattice units. The spacing between fibres in orthogonal planes does not have to be the same but, in a given plane, that spacing is kept constant. The bonds between nodes are assigned different elastic constants for the matrix and for the fibre.

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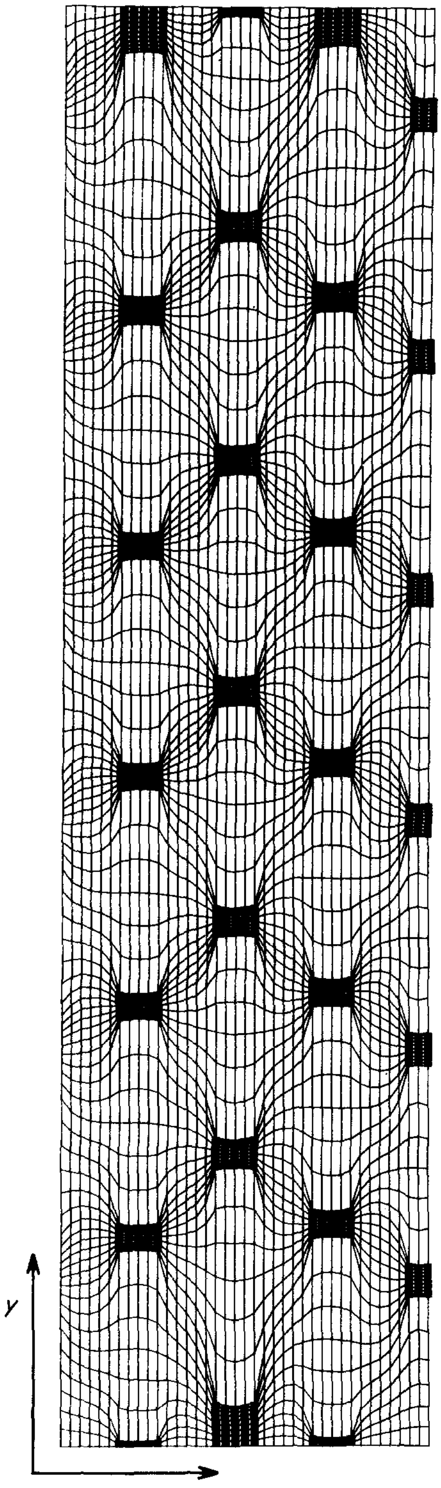

Figure 2 Two-dimensional representation of the deformation scheme in the x–y plane for a fibre reinforced composite with $l/d = 2$, $E_f/E_m = 20$. The fibre volume fraction V_f was set equal to 0.3. The x and y scales have been distorted so that the stress distribution can be easily seen.

neighbours, by a systematic sequence of operations, which steadily reduces the net residual force acting on each node.

That approach [3], applied to single-fibre reinforced composites, is now extended to multi-fibre composites. The dependence of the composite modulus on the aspect ratio and volume fraction of the fibres is studied and the predictions are compared to those of the widely used semi-empirical Halpin–Tsai equation [4]. Our results are also compared to available experimental data on the stiffness of foams and particulate-reinforced composites. We find a much better agreement with experiment than that obtained using the Halpin–Tsai equation. Application of our model to a detailed study of the stress concentration around the fibre ends, and of the effect of fibre-matrix adhesion on the composite modulus, will be presented in a forthcoming publication.

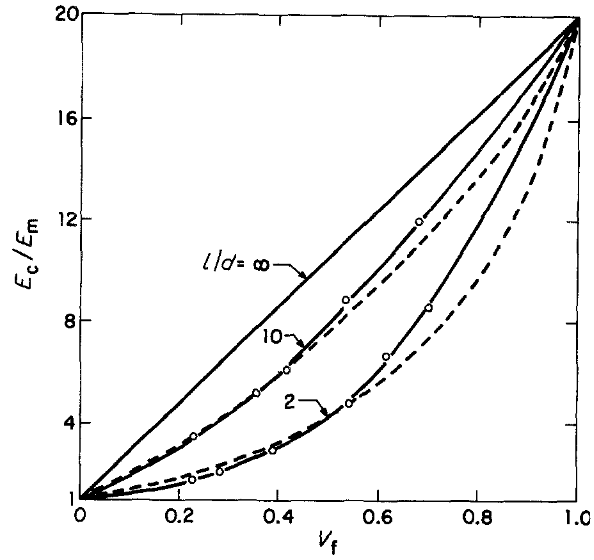

Figure 3 Dependence of the composite modulus E_c (in units of E_m) on the volume fraction V_f of the fibres for a ratio $E_f/E_m = 20$. The curves are for different values of the fibre aspect ratio l/d. The fibre diameter d was set to values ranging from 3 lattice units (at $V_f = 0.2$) to 9 units (at $V_f = 0.7$). We took $\nu_m = \nu_f = 0.3$. The dashed line gives the Halpin–Tsai [4] prediction.

2. Model

We start by describing our three-dimensional simple cubic model for a unidirectional multi-fibre composite. A two-dimensional representation (Figs 1a and b) is given of the lattice model in a plane parallel (Fig. 1a)

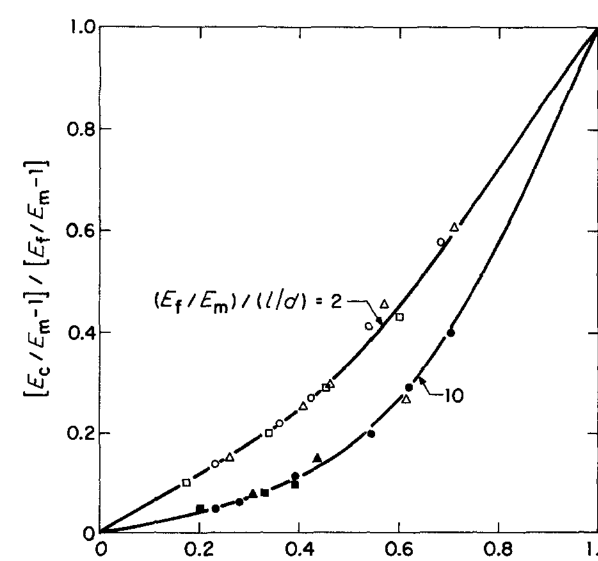

Figure 4 Composition dependence of the modulus E_c (suitably normalized) for different values of the ratio $(E_f/E_m)/(l/d)$. Values for the pairs $(E_f/E_m, l/d)$ are as follows: (○) (20, 10); (□) (6.7, 3.3); (△) (60, 30); (●) (20, 2); (■) (60, 6); (▲) (10, 1). We took $\nu_m = \nu_f = 0.3$.

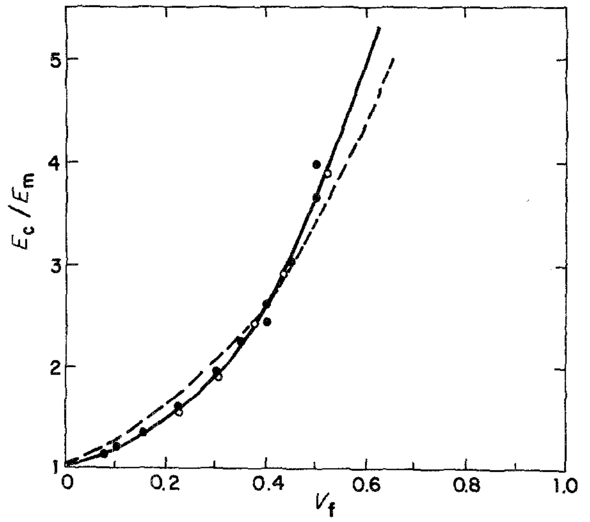

Figure 5 Composition dependence of E_c/E_m for glass spheres in epoxy. O: experiments [7]; (–O–) present model. Values for the parameters are as follows: $E_f/E_m = 25$, $\nu_f = 0.23$, $\nu_m = 0.39$. The dashed line indicates the Halpin–Tsai prediction.

and perpendicular (Fig. 1b), respectively, to the direction of loading (y-direction). The lattice comprises 120 nodes along the y-axis and 39 in the x- and z-directions. The parameters d and l denote the fibre diameter and length, respectively. In the simulations, d was set equal to 3 to 7 lattice units. The spacing between fibres in orthogonal planes does not have to be the same but, in a given plane, that spacing is kept constant. The bonds between nodes are assigned different elastic constants for the matrix and for the fibre. The Young and shear moduli of the matrix and of the fibre are denoted by (E_m, G_m) and (E_f, G_f), respectively. For simplicity, we assume both components to be isotropic so that $G_m = E_m/2(1 + \nu_m)$ and $G_f = E_f/2(1 + \nu_f)$, where ν denotes a Poisson ratio.

The composite lattice is strained by a constant amount (1%) along the y-axis. In order to estimate the relative displacements of the fibre and the matrix, each node is relaxed towards local mechanical equilibrium with its neighbours. This is done with the help of a sequence of fast computer algorithms, described previously [3], which steadily reduces the net residual

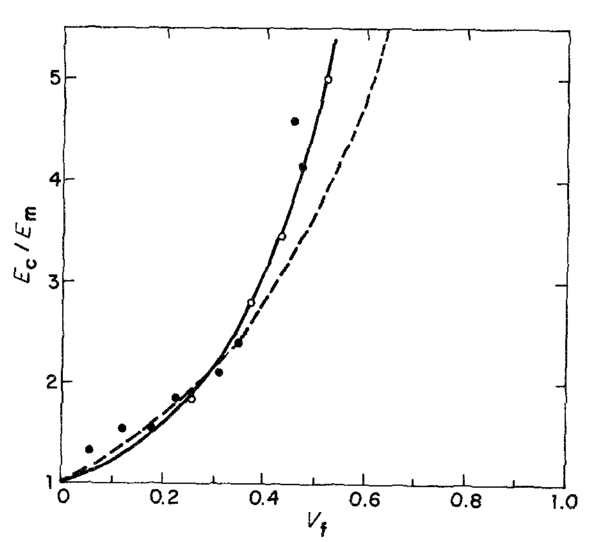

Figure 6 As Fig. 5, but for glass fibres in polyester resin. Experimental data [8]. $E_f/E_m = 42$, $\nu_f = 0.21$, $\nu_m = 0.44$.

force acting on each node. For simplicity, we assume that the displacements of the nodes along the coordinates axes are mutually independent and we focus on motions along the y-axis. Since the shearing mode along the y-axis predominates when it comes to load transfer between the matrix and the fibres, lateral motions in the x- and z-directions should be of secondary importance for the present study [3–5]. The validity of our assumption has also been discussed [3].

Fig. 2 gives a two-dimensional representation of the deformation scheme in the $x-y$ plane for a fibre reinforced composite with $l/d = 2$, $E_f/E_m = 20$. The fibre volume fraction V_f was set equal to 0.3. The x and y scales have been distorted so that the stress distribution can be easily seen. The figure shows a high degree of shearing in the matrix between nearest neighbour fibres along the transverse x-axis, indicative of the importance of the shearing mode in the load transfer process between adjacent fibres.

3. Results and discussion

The dependence of the composite modulus E_c (in units of E_m) on the volume fraction V_f of the fibre for a ratio $E_f/E_m = 20$ is shown in Fig. 3. The curves are for different values of the fibre aspect ratio l/d. For continuous fibres ($l/d = \infty$), the dependence is linear and E_c reaches its maximum value given by the so-called Voigt average $E_c = E_f V_f + E_m(1 - V_f)$. For discontinuous fibres, the curves show a negative deviation from the ideal continuous case and the modulus decreases with a decrease in the aspect ratio. The results in Fig. 3 have also been compared to the predictions of the widely used semi-empirical Halpin–Tsai equation [4]. Our values are seen to be lower for $V_f < 0.5$ and higher for $V_f > 0.5$, the discrepancy between the two sets of predictions being larger for lower values of the aspect ratio.

The results presented above thus indicate that, for a given value of V_f, the composite modulus increases with an increase in the fibre aspect ratio l/d and in the fibre modulus E_f (for given E_m). It seems therefore natural to inquire whether these two fibre characteristics can be unified in any way. Fig. 4 depicts the composition dependence of E_c (suitably normalized) for two different values of the ratio $(E_f/E_m)/(l/d)$. Although E_f/E_m values within a given ratio vary by a factor as large as 10, all our results for a given $(E_f/E_m)/(l/d)$ fall on the same curve. That ratio [6] thus seems to be an important index of fibre reinforced composites: the higher the ratio, the higher the efficiency of load transfer between the matrix and the fibre.

The predictions of our model to experimental data on several particulate ($l/d = 1$) filled polymers are compared in Figs 5 to 7: glass spheres in thermosetting resins [7, 8] and natural silica particles in epoxy [9]. Our results are in good quantitative agreement with experiment. The Halpin–Tsai equation [4], on the other hand, is seen to systematically underestimate the composite modulus at high volume fractions of the filler.

The results presented above were for filler particles which are stiffer than the surrounding matrix. We now turn to application to materials, like certain polyblends

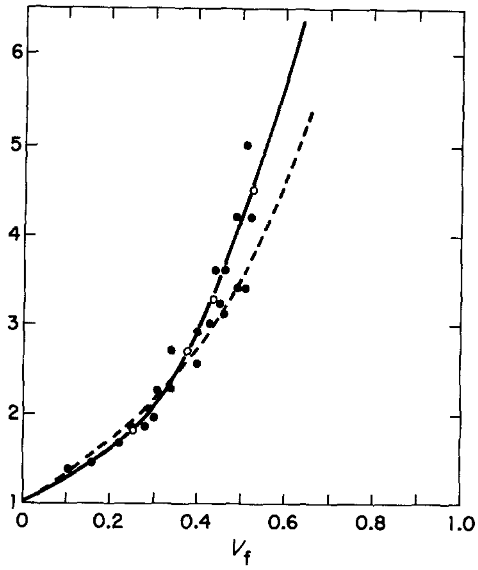

Figure 7 As Fig. 5, but for silica particles in epoxy. Experimental data [9]. $E_f/E_m = 35$, $v_f = 0.25$, $v_m = 0.4$.

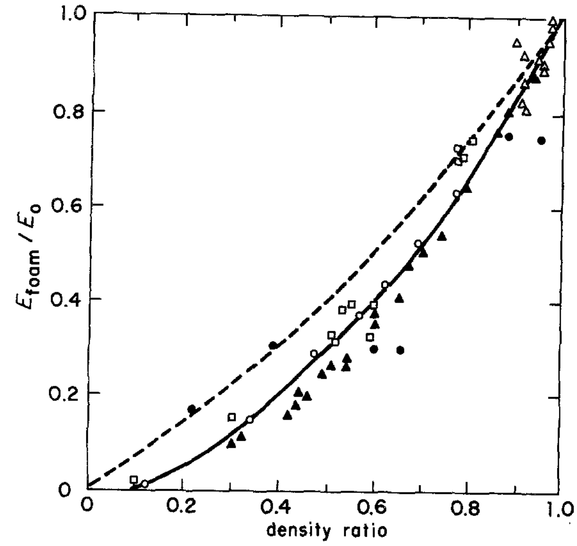

Figure 8 Dependence of the modulus on the density ratio for thermoplastic foams. (–○–) present model. The other symbols are for experimental data: (□) [10]; (●) [11]; (▲) [12]; (△) [13]. The dashed line indicates the Halpin–Tsai prediction.

and polymeric foams, in which the continuous phase is more rigid than the included phase. Fig. 8 compares our results for the modulus to experimental data on several thermoplastic foams [10–13]. We took $l/d = 1$ for the included phase. The results here follow the often conjectured [1, 13] density-squared relationship. Again, the agreement with experiment is excellent and far better than that obtained with the help of the widely used Halpin–Tsai equation [4]. We have also studied the dependence of our results of Fig. 8 on the distribution and size of the included phase. As far as we could determine, the modulus of the foam seems to be a unique function of the average density ratio.

To conclude, we have presented a finite-difference type of approach for the study of the elastic properties of short fibre and particulate-filled polymers. Our results are in better quantitative agreement with experiment than those obtained with the help of the semi-empirical Halpin–Tsai equation. Another major advantage over previous approaches [1, 4] is that the present model is microscopic in nature and allows a study of the effect of the fibre size distribution and of the fibre–matrix adhesion [3] on the composite modulus. The model also permits an easy evaluation of the stress distribution inside the material and therefore lends itself to a detailed study of the fracture process itself [14, 15]. These studies will be the object of a forthcoming publication.

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