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ORIGINAL CONTRIBUTION

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Theoretical and experimental investigation of the stiffness of iron-filled epoxy polymers

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Abstract The static elastic moduli of particulate-filled epoxy resins, consisting of two phases, one of which has isotropic-elastic and the other linear viscoelastic properties, were studied. The effects of parameters such as the filler volume fraction, the filler size and aspect ratio, and the filler distribution were evaluated. For this purpose, in the theoretical part a development of a simplified model based on mechanics of materials approach was used. In the experimental part a class of iron-filled epoxy composites with various filler content, filler size, and filler distribution was subjected to tests in order to obtain the elastic modulus. The experimental results were compared with the theoretical values derived from the developed model as well as with theoretical values obtained from other investigators.

Key words Composite material – filler size and aspect ratio – filler volume fraction – filler-matrix adhesion

Introduction

The introduction of metal particles into polymeric matrices results in composites which are characterized by enhanced mechanical properties, such as their moduli and their fracture toughness. In contrast, addition of metal particles to polymeric matrices can produce a reduction in thermal properties such as their thermal expansion coefficient.

A rigorous description of a composite system consisting of a matrix, in which filler particles have been dispersed, is not an easy task. In fact, a great number of geometrical, topological, mechanical, etc. parameters are necessary, the majority of which varies statistically or is simply unknown. Theoretical treatments usually attempt to exploit, as much as possible, readily available information, which, in most cases, consists of the mechanical properties of the matrix and the filler and the volume fraction of the latter, while suitable assumptions cover missing data.

However, one of the main problems remains the prediction of the composite properties when the properties of the constituent materials are known. The difficulty of this problem arises from the fact that the thermomechanical properties of a composite depend on a large number of parameters, such as the individual properties of the filler and the matrix, the shape and size of the filler, the filler volume fraction, the filler aspect ratio, the filler distribution, and the resin-filler adhesion etc.

Analytical solutions are valid up to some fairly low filler-volume fraction, as they have to ignore, for reasons of efficiency, any mechanical interaction between neighboring inclusions. Referring in particular to the moduli, a great number of empirical or semiempirical expressions exist which either express a kind of law of mixtures, or are simply an attempt to match theoretical curves to experimental data. In most of them a perfect adhesion between matrix and filler was assumed as existing between the phases of the composite.

The various theoretical models that have been proposed [1–24] to predict the mechanical properties of filled composites have emphasized particular parameters. The filler volume fraction and the mode of packing were the parameters studied in the models presented in refs. [1–3, 12–14], while the importance of the filler size on the final properties of composites was discussed in [4–6, 8, 16, 17]. The effect of the filler-matrix adhesion on the elastic moduli of composites has been discussed in refs. [16, 17, 22–24]. Recently, a large survey by Ahmed and Jones [25] was presented of the existing theories for predicting the elastic modulus and the strength of particulate-filled polymeric composites.

In the present investigation the elastic moduli of particulate-filled polymers were studied. The effects of parameters such as the filler volume fraction, the filler size and aspect ratio, the filler distribution were evaluated. In the theoretical part of the work a development of a simplified model based on mechanics of materials approach was used. In the experimental part, a class of iron-filled epoxy composites with various filler volume fraction, size and distribution was subjected to tests in order to evaluate the elastic modulus. Finally, a comparison of the experimental results with the theoretical values obtained from the present model and from other authors was made.

Theoretical and semi-empirical formula

A large number of theoretical and semi-empirical expressions for the effective moduli exist in the literature. Some of them are derived from the theory of elasticity, whereas others express some kindof law of mixtures or try to match thoretical expressions to experimental data by appropriately defining the existing constants in these expressions.

One of the earliest theories for a composite system was developed for elastomers and is based on Einstein's equation for the viscosity of a suspension of rigid spherical inclusions [26]:

$$\frac{E_c}{E_m} = 1 + au_f \,, \tag{1}$$

where E_c, E_m are the elastic moduli of the composite and matrix and u_f is the filler volume fraction. The constant a is equal either to 2.5 or 1. Next, according to an equation developed by Guth and Smallwood [27–28] which is an extension of the Einstein equation, it follows that

$$\frac{E_c}{E_m} = 1 + 2.5u_f + 14.1u_f^2 \,. \tag{2}$$

An equation based on a mathematical model valid for the glassy behavior of composites is from Kerner [2]; for rigid fillers it simplifies to the expression:

$$\frac{E_c}{e_M} = 1 + \frac{u_f}{u_m}\left[\frac{15(1-v_m)}{8-10v_m}\right], \tag{3}$$

where v_m is the Poisson ratio of the matrix.

A relation taking into account the effect of adhesion efficiency between the two phases has been suggested by Sato and Furukawa [29] and is expressed by

$$E_c = E_m\left[\left[1 + \frac{1}{2}\frac{Y^2}{1-Y}\right]\cdot\left[1 - \frac{Y^3T}{3}\left(\frac{1+Y-Y^2}{1-Y+Y^2}\right)\right] - \frac{Y^2T}{3(1-Y)}\left(\frac{1+Y-Y^2}{1-Y+Y^2}\right)\right], \tag{4}$$

where $Y = u_f^{1/3}$ and T is and adhesion factor, taking the value of zero for perfect adhesion and the value of 1 for zero adhesion.

On the other hand, the Mooney equation [30] can take into consideration a number of effects of the filler agglomeration:

$$\frac{E_c}{E_m} = \exp\left(\frac{2.5u_f}{1-su_f}\right) \tag{5}$$

by means of a crowding factor s, expressing the ratio of the apparent volume occupied by the filler over its own true volume. This factor takes values from 1 to 2, depending on the type of particle distribution in the matrix material. For closely packed spheres of a uniform size, this is $s = 1.35$.

In the equation proposed by Eilers and Van Dyk [31],

$$\frac{E_c}{E_m} = \left(1 + \frac{ku_f}{1-S'u_f}\right), \tag{6}$$

k and S' are constants usually equal to 1.25 and 1.20, respectively. The effect of filler concentration on the elastic modulus is also expressed by an empirical relation proposed by Bills et al. [32] which is written as

$$\frac{E_c}{E_m} = \exp[Au_f/(1-Bu_f)] \,, \tag{7}$$

where A and B are experimental constants. They have found that constant A takes the value 2.5 while B is given by

$$B = -6.4 \times 10^{-3}T + 2.51$$

in which T corresponds to the test temperature.

A semiempirical single-parameter equation describing the moduli of particulate systems has been formulated by Narkis [33] as follows:

$$\frac{E_c}{E_m} = 1/[K(1-u_f^{1/3})] \,, \tag{8}$$

where K is an empirical parameter related to a stress concentration factor with usual values in the range of 1.4–1.7.

Analytical equations for the elastic modulus of a composite containing spherical fillers have also been derived by Takahashi et al. [34]. In the case of perfect adhesion they gave the relationship

$$\frac{E_c}{E_m} = 1 + (1 - v_m)\left[\frac{E_f(1 - 2v_m) - E_m(1 - v_f)}{E_f(1 + v_m) + 2E_m(1 - 2v_f)} + \frac{10(1 + v_m)E_f(1 + v_m) - E_m(1 + v_f)}{2E_f(4 - 5v_m)(1 + v_m) + E_m(7 - 5v_m)(1 + v_f)}\right]u_f , \tag{9}$$

and in the case of imperfect adhesion

$$\frac{E_c}{E_m} = 1 + (1 - v_m)\left[\frac{E_f(1 - 2v_m) - E_m(1 - v_f)}{E_f(1 + v_m) + 2E_m(1 - 2v_f)} + \frac{10(1 + v_m)E_f(1 + v_m)(7 + 5v_f) - 4E_m(1 + v_f)(7 - 4v_f)u_f}{E_f(1 + v_m)(17 - 19v_m)(7 + 5v_f) + 4E_m(7 - 5v_m)(1 + v_f)(7 - 4v_f)}\right]u_f . \tag{10}$$

Halpin and Tsai [35] developed an interpolation procedure that is an approximate representation of more complicated micromechanics results. They give the following relationship for the transverse elastic modulus of composites reinforced with unidirectional continuous or short fibers.

$$E_c = E_m \frac{1 + \xi \eta u_f}{1 - \eta u_f} \tag{11}$$

with

$$\eta = \frac{E_f - E_m}{E_f + \xi E_m}$$

and

$$\xi = 1 + 40 u_f^{10} .$$

Finally, a lower bound for the elastic modulus of unidirectional composite materials which is valid for E_c in transverse direction is given by the relationship

$$\frac{1}{E_c} = \frac{u_f}{E_f} + \frac{u_m}{E_m} . \tag{12}$$

Theoretical analysis

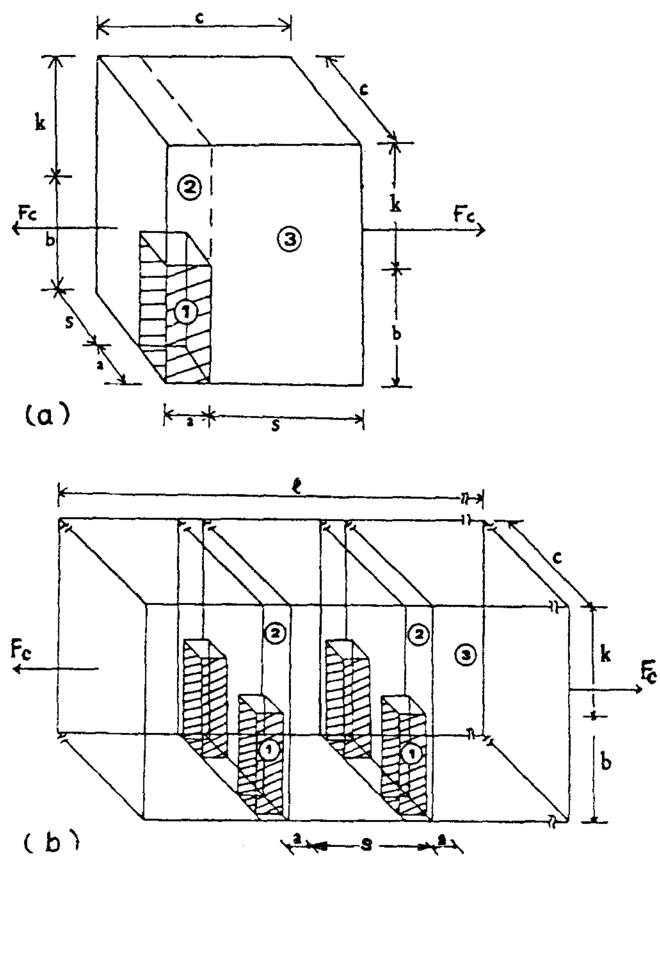
Since there is a need for a proved theoretical model of reinforced polymers which would make possible to predict their stiffness with a reasonable degree of confidence, the "prism-within-prism" model introduced by Ogorkiewicz and Weidmann [36] was improved, by considering inclusions such as short fibers connected in series and in parallel with matrix material. From this model, where short fibers rather than spheres are involved, more realistic results might be expected. This model consists of rectangular prisms of the filler material, with their long axis perpendicular to the direction of filler alignment and the applied tensile load, within a prism of the polymer matrix. The dimensions of the prisms are fixed by the dimensions of the specimens used in the tests in order to make a better comparison between theory and experiment (see Fig. 1). As shown in the Appendix, such a "prism-within-prism" model leads to the following equations for the elastic modulus E_{ct}.

For a single prismatic inclusion:

$$E_{ct} = \frac{E_m \cdot [E_f \cdot a \cdot b \cdot c + E_m \cdot [(b + k) \cdot c - a \cdot b] \cdot c]}{E_f \cdot a \cdot b \cdot s + E_m \cdot [a \cdot c \cdot (b + k) + s \cdot [(b + k) \cdot c - a \cdot b]]} , \tag{13}$$

with the filler volume fraction being $u_f = \frac{a^2 \cdot b}{c^2(b + k)}$.

Fig. 1A, B Representative elements of the theoretical model used



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In the above relationships a, b are the dimensions of the inclusions, s and k are the longitudinal (parallel to the load F_c) and perpendicular side-to-side spacing of the inclusions respectively, c is the side of the matrix-inclusion prism denoting the representative element of the composite material, and E_f, E_m are the elastic moduli of filler and matrix respectively.

For n prismatic inclusions in series and v prismatic inclusions in parallel connection with matrix material

$$E_{ct} = \frac{E_m \cdot [E_f \cdot v \cdot a \cdot b \cdot l + l \cdot E_m[(b+k) \cdot c - v \cdot a \cdot b]}{E_f \cdot v \cdot a \cdot b(l - n \cdot a) + E_m[n \cdot a \cdot (b+k) \cdot c + (l - n \cdot a)[(b+k) \cdot c - v \cdot a \cdot b]]} \quad (14)$$

with the filler volume fraction being $u_f = \dfrac{v \cdot n \cdot a^2 \cdot b}{c \cdot l \cdot (b+k)}$

In the above relationships l and c are the sides of the matrix-inclusions prism denoting the representative element of the composite material.

Experimental work

In order to verify the theoretical expressions given by Eqs. (13, 14), experiments were carried out with metalic inclusions having cylindrical shape in order to avoid the stress singularities arising in the corners of prismatic inclusions. These inclusions were polished before use to remove any sharpness and to improve the adhesion with the matrix. The matrix material was in all cases a cold-setting system based on a diglycidil ether of bisphenol-A resin together with plasticizer 20% by weight of resin cured with 8% triethylenetetramine. The composites were manufactured in the strength of Materials Laboratory of the Technical University of Athens. The elastic modulus and Poisson ratio of the constituent materials, which were determined through tensile tests by using strain-gauges are as follows:

Iron inclusions: $E_f = 210 \times 10^9 \mathrm{N/m^2}$ and $v_f = 0.29$.
Epoxy resin matrix: $E_m = 3.18 \times 10^9 \mathrm{N/m^2}$, $v_m = 0.37$.

Dogbone specimens with dimensions 165 mm, 29 mm, 9 mm for length, width and thickness, respectively, were used during the tests. The dimensions at the measuring area were 50 mm, 19 mm, 9 mm for length, width and thickness respectively (see Figs. 2, 3).

Tensile measurements were carried out in a conventional Instron-type testing machine at room temperature with a rate of extension $c = 0.2$ mm/sec. Three series of specimens with inclusion diameter $d = 4$ mm, 5.2 mm (two rows of inclusions) and $d = 8$ mm (one row) were used (see Figs. 2, 3) in order to study the effect of filler-volume fraction on the elastic modulus of composites. For the strain measurement, strain-gauges (KYOWA type KFG-5-120-D16, $k = 1.99$) were placed on the specimens.

Four specimens per each case were used during the experiments. The results presented for E_c are the arithmetic mean.

Results and discussion

The families of strain-stress curves of iron-epoxy composites as obtained from the experiments are shown in Figs. 4–6 for filler diameter $d = 4$ mm, 5.2 mm, and 8 mm,

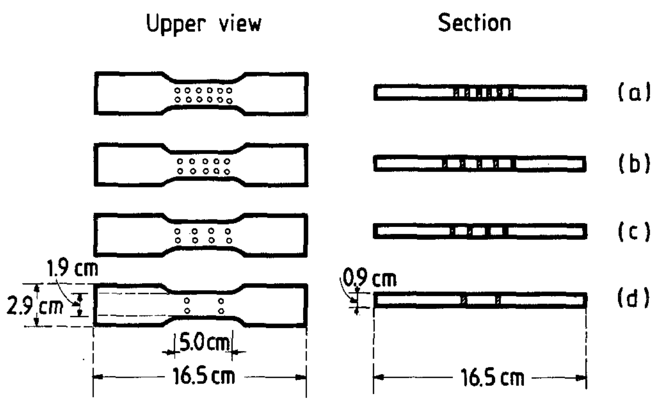
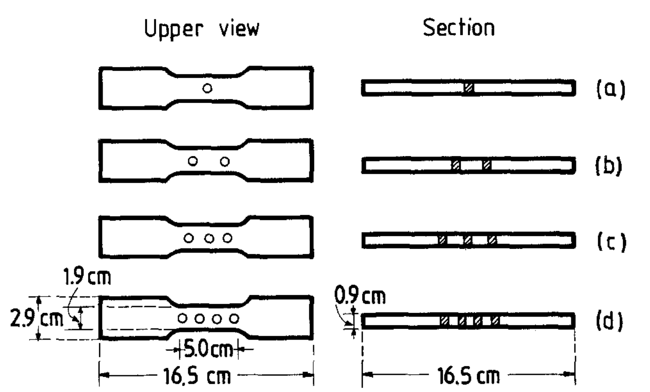


Fig. 2 Specimens of epoxy resin reinforced with cylindrical inclusions having diameter $d = 4$ mm and $d = 5.2$ mm

Fig. 3 Specimens of epoxy resin reinforced with cylindrical inclusions having diameter $d = 8$ mm



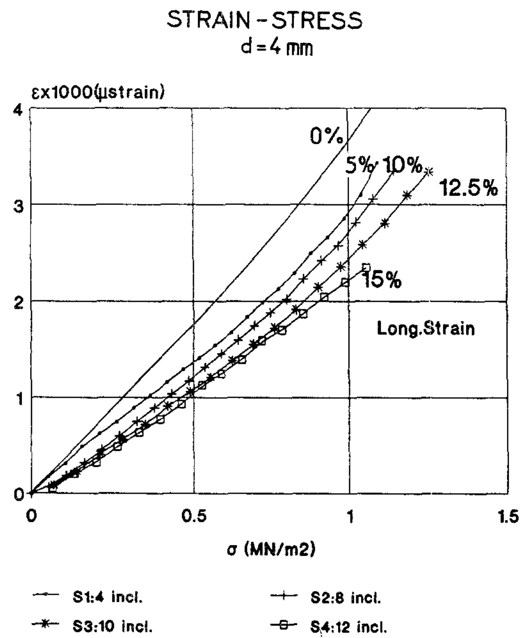


Fig. 4 Strain-stress curves obtained from experiments with specimens of epoxy resin reinforced with cylindrical inclusions having diameter $d = 4$ mm, for various filler volume fractions

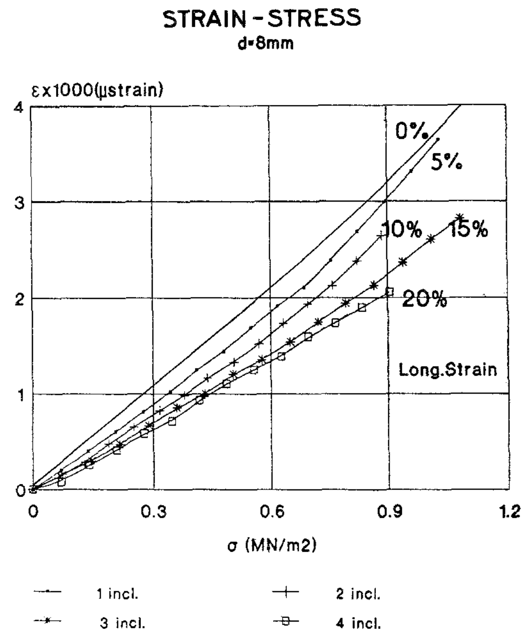


Fig. 6 Strain-stress curves obtained from experiments with specimens of epoxy resin reinforced with cylindrical inclusions having diameter $d = 8$ mm, for various filler volume fractions

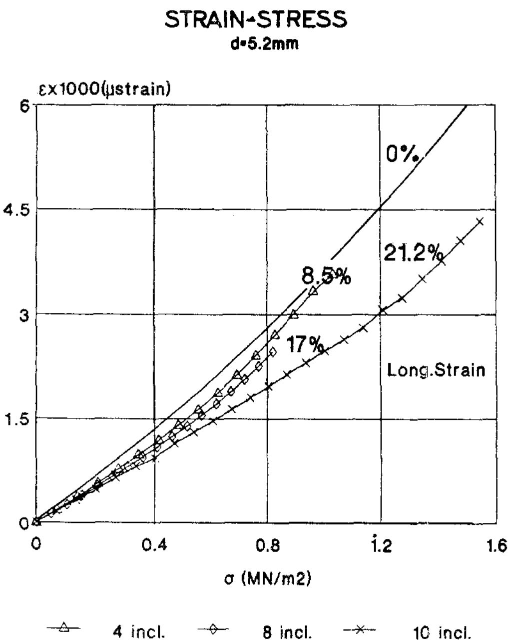


Fig. 5 Strain-stress curves obtained from experiments with specimens of epoxy resin reinforced with cylindrical inclusions having diameter $d = 5.2$ mm, for various filler volume fractions

respectively. From these curves the effect of filler-volume fraction on the mechanical behavior of these composites becomes clear during the entire range of filler diameters. From the form of these curves it can be deduced that the gross behavior of the composites exhibits an obvious degree of nonlinearity. Because of the stress and strain-concentrations, which exist in local regions beween fillers, the elastic limit of the matrix material may be exceeded long before the gross behavior of the composite exhibits the nonlinear response. On the other hand, in our case where the matrix material used is a viscoelastic material, the nonlinear response may be due, at least in part, to viscoelastic, i.e., time-dependent effects. Moreover, as the applied loading exceeds the yield point of the composite, the material may exhibit a nonlinear time-independent deformational response. It is obvious that an elastic analysis is applicable only in the initial portion of the stress-strain response.

From the same curves it can be deduced that, as the filler-volume fraction is increased, a more linear elastic response is obtained. This kind of behavior is expected since the filler material is strongly elastic, so that, as the filler-volume fraction is increased the viscoelastic response of the composite is decreased.

In Fig. 7 the elastic modulus of the composites, E_c, as obtained from the experiments is plotted versus filler-volume fraction, u_f, for the three filler diameters. As expected, the modulus increases with increasing values of u_f. Also, it can be observed that for filler diameter $d = 4.2$ mm higher values of elastic modulus were obtained.

Figures 8a, b, c show the theoretical values of the elastic modulus, E_c, of the composite as calculated by Eq. (14)

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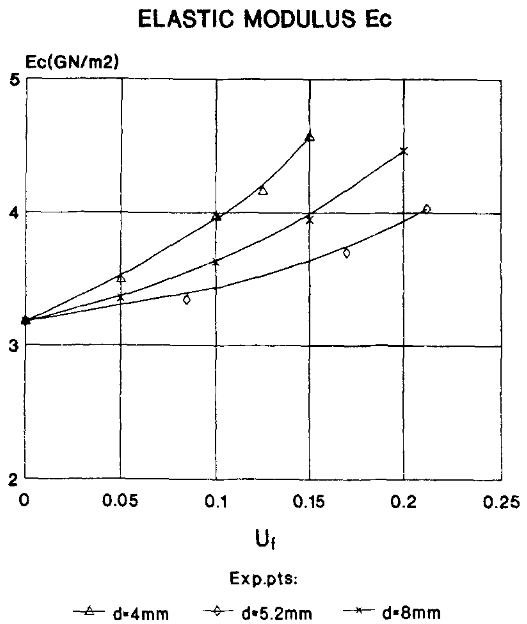


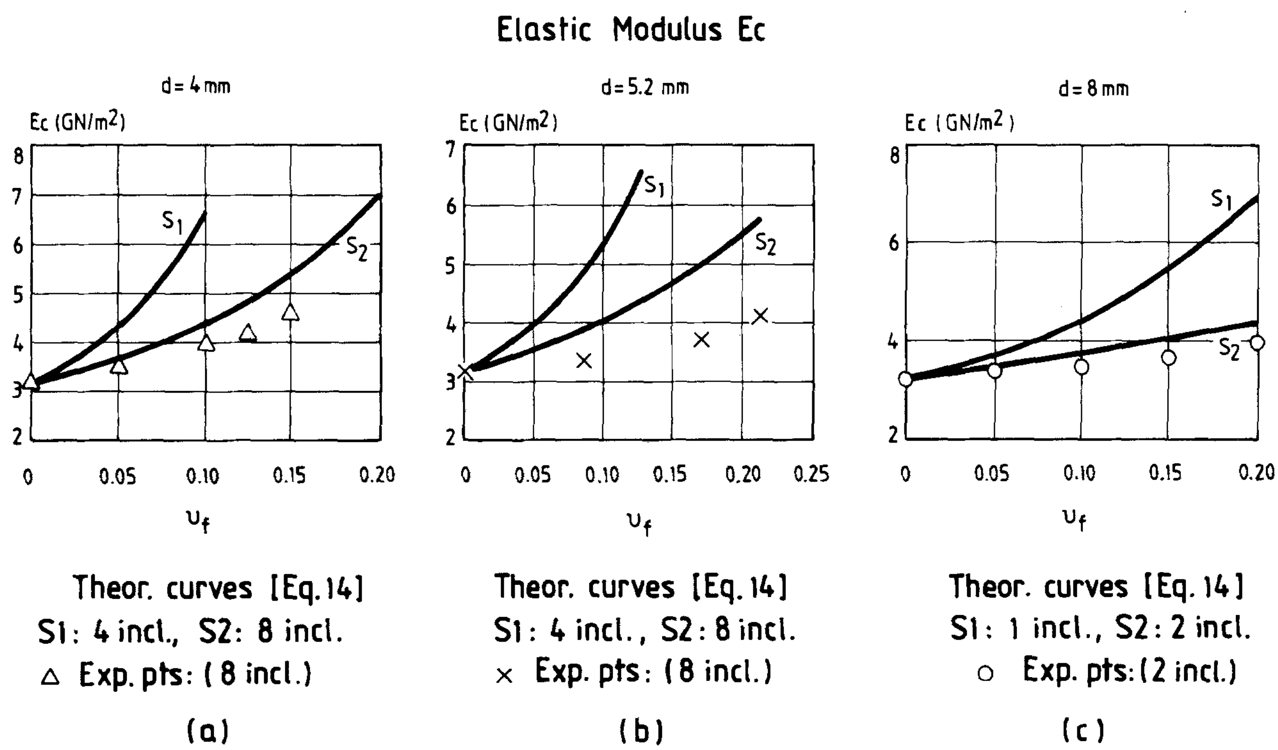
Fig. 7 Experimental values of the elastic modulus versus filler volume fraction for filler diameters $d = 4, 5.2, 8$ mm

versus the filler-volume fraction for the three filler diameters. In the same figures the experimental results are also plotted. It can be observed that the theoretical values are always above the experimental ones and that the difference increases with increasing volume fraction. Also, for filler diameter $d = 5.2$ mm where the lowest experimental values of E_c were obtained the discrepancies between theory and experiment are larger than in the other two cases.

In Figs. 9a, b, c the experimentally obtained elastic moduli of the composite were plotted versus filler-volume fraction u_f together with the theoretical curves given by Eqs. (1–12) and obtained by other investigators in refs. [26–35] for comparison. From this figure it can be observed that the experimental results for filler diameter $d = 4$ mm and 8 mm fit well with the theoretical results given by Kerner (Eq. (3)), Einstein (Eq. (1)), Sato–Furukawa ($T = 0$ (Eq. (4)), Halpin–Tsai (Eq. (11)), and Eilers Van Dyck (Eq. (6)). On the other hand, the experimental results for $d = 5.2$ mm fit well with the theoretical results given by Einstein (Eq. (1)), Takahashi (Eq. (10)), and with the lower bound (Eq. (12)). This may mean that the discrepancies between the experimental results and the theoretical values obtained by the model applied in this paper are instead due to the fact that this model gives high values. Indeed, it is a model based on the mechanics of material approach which is not so realistic and so accurate as the theory of elasticity.

Moreover, factors such as adhesion efficiency at the interface on the two phases, which play an important role, are not taken into account in this model. This is a decisive

Fig. 8A, B, C Theoretical values of the elastic modulus versus filler volume fraction for filler diameter $d = 5, 5.2$ and 8 mm respectively



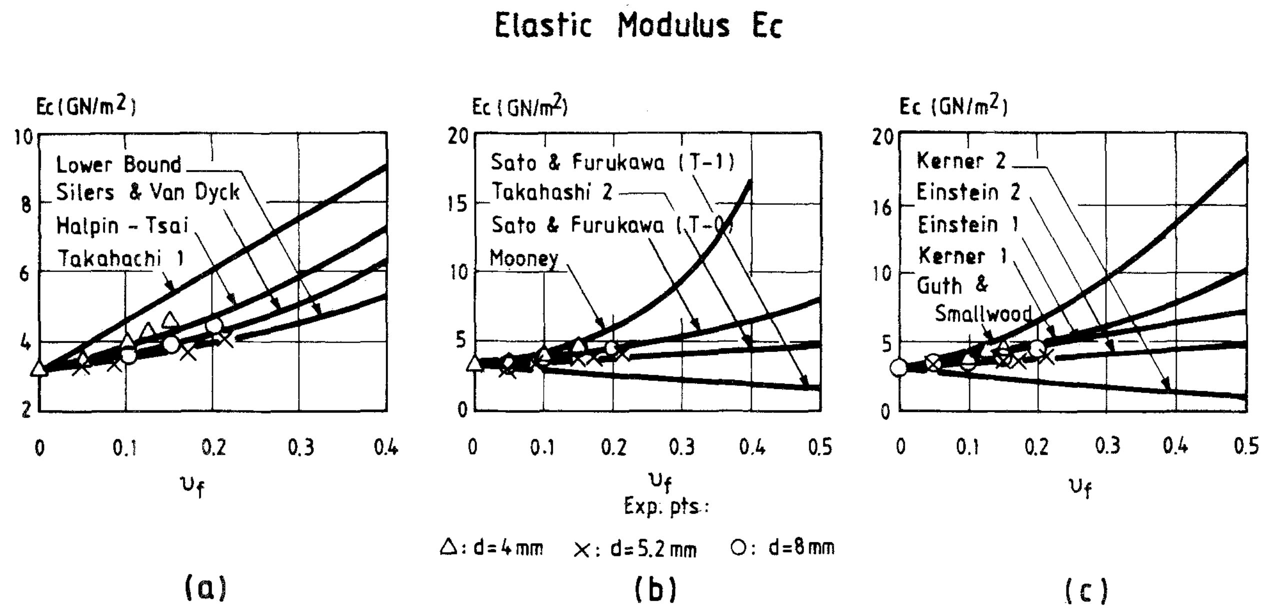


Fig. 9A, B, C Experimental values of the elastic modulus of the composite versus filler volume fraction compared with the theoretical curves

factor for the behavior of the composite. In most theoretical models this adhesion is considered as perfect, i.e., the interface can ensure continuity of stresses and displacements. However, such a condition is hardly fulfilled in real composites. In reality, around an inclusion embedded in a matrix a rather complex situation develops, consisting of areas of imperfect bonding, mechanical stresses, due to shrinkage, or even stress singularities, due to the geometry of the inclusion. These factors, which influence the adhesion efficiency, become greater when filler volume fraction is increased. Thus, a theory which does not take this fact into account may present more discrepancies with experimental results. The use of a model taking into account the adhesion between inclusions and matrix can be a next step in the theoretical part of this research.

On the other hand, no special care was taken during the manufacturing of the specimens (except polishing the inclusions) to improve the adhesion between filler and the polymer matrix. The use of an agent to improve the adhesion between inclusions and matrix can be a next step in the experimental part of this research in order to make comparisons with the present case.

An other possible reason – perhaps the main reason – for the discrepancies between theory and experiment are the shape and the dimensions of the inclusions. The majority of the theoretical and semi-empirical formulae of the literature for the elastic modulus are given for composites reinforced with spherical inclusions having diameter much less than 1 mm, which represents a real material. Our experiments were carried out with specimens of polymer composite reinforced with cylindrical inclusions having diameter 4–8 mm and a given distribution, i.e., specimens which serve as a model rather than a real material.

Finally, the fact that the influence from boundaries has not been considered, something which normally should not have been neglected with these short inclusions, can be a reason for discrepancies.

Conclusions

In the present paper a theoretical model predicting the elastic modulus of a filled polymer was applied and a series of factors such as filler diameter, filler distribution, and filler volume fraction affecting the mechanical behavior of iron-epoxy composites were investigated. A comparison between theoretical predictions given by the present model as well as by other authors with experimental results was made.

From this investigation the following conclusions may be derived:

1) The gross-behavior of the composites under investigation exhibits an obvious degree of non-linearity, while as the filler-volume fraction is increased a more linear response is obtained.
2) The variation of the diameter, d, and thus of the aspect ratio of the inclusions affects the elastic modulus.
3) The experimental results present discrepancies with the theoretical ones obtained by the investigated model especially for $d = 5.2$ mm.

4) The experimental results fit well with some other theoretical predictions given in the literature.
5) The results of the applied theoretical model are in good agreement with the results of the most known theoretical predictions of the literature. But, this is possibly due to the low filler volume fraction.

Appendix

Basic model

According to Ogorkiewicz and Weidmann approach [36] a composite consisting of a polymeric matrix containing unidirectionally aligned discontinuous fibers may be idealized into a prism of the polymeric material and within it, a prism of fiber material. The volume of the latter, expressed as a fraction of the volume of the larger prism, is equal to the volume fraction of the fibers and its proportions are fixed by the aspect ratio of the fibers. The proportions of the outer prism are more difficult to define because they are related to the spacing of the fibers, which is not generally known. Therefore, assumptions have to be made about it, and the authors [36] have assumed that the end-to-end and side-to-side spacings of fibers are equal. Such an assumption about the equality of spacings appears reasonable on physical grounds, particualrly when the fibers are relatively short, as well as being mathematically tractable, and leads to the model shown in Fig. (1a).

The theoretical approach is based on the following assumptions:

i) The composite material is linearly-elastic, that is, Hooke's law is valid.
ii) Filler distribution is uniform, so that the composite may be regarded as a quasihomogeneous isotropic material.
iii) The matrix and the fillers are elastic, isotropic, and homogeneous.

Under the action of a tensile load F_c acting perpendicularly to the long sides of the inclusion prism, the model shown in Fig. (1a) may be considered to consist of two parts. One of these parts consists of the inclusion prism (1), with cross-sectional area $a \cdot b$, in parallel with a matrix prism (2). The other part consists of the remainder of the matrix material (3) in series with (1) + (2). In Fig. 1a $s = c - a$ denotes the side-to-side spacing of the inclusions and k a parameter depending on the inclusion length which becomes zero when the inclusion is continuous.

Equilibrium of forces in the direction of the applied uniaxial tensile stress σ_c gives the following equation, where the subscripts 1–3 refer to the different layers, or parts of layers of the model:

$$\sigma_c \cdot (b + k) \cdot c = \sigma_3 (b + k) \cdot c \Rightarrow \sigma_c = \sigma_3 \tag{A1}$$

$$\sigma_c (b + k) \cdot c = \sigma_1 \cdot a \cdot b + \sigma_2 [(b + k) \cdot c - a \cdot b] \tag{A2}$$

If it is assumed that there is "perfect bonding" between the inclusion and the matrix, then:

$$\varepsilon_1 = \varepsilon_2 \tag{A3}$$

and from the geometry of the model the total extension is

$$\varepsilon_c \cdot c = \varepsilon_c (s + a) = \varepsilon_1 \cdot a + \varepsilon_3 \cdot s \,. \tag{A4}$$

The constitutive equations, assuming proportionality of stress to strain are:

$$\sigma_c = E_{ct} \cdot \varepsilon_c \tag{A5}$$

$$\sigma_1 = E_f \cdot \varepsilon_1 \tag{A6}$$

$$\sigma_2 = E_m \cdot \varepsilon_2 \tag{A7}$$

$$\sigma_3 = E_m \cdot \varepsilon_3 , \tag{A8}$$

where E_{ct}, E_f, E_m are the elastic moduli of the composite, inclusion, and the matrix, respectively. Also, $\sigma_1, \sigma_2, \sigma_3$ and $\varepsilon_1, \varepsilon_2, \varepsilon_3$ are the stress and strain of the layers (1), (2) and (3), respectively.

Combination of Eqs. (A2), (A5), (A6), (A7) gives:

$$E_{ct} \cdot \varepsilon_c (b + k) \cdot c = E_f \cdot \varepsilon_1 \cdot a \cdot b + E_m \cdot \varepsilon_2 [(b + k) \cdot c - a \cdot b] \tag{A9}$$

From Eqs. (A1), (A5), (A10), we have

$$E_{ct} \cdot \varepsilon_c = E_m \cdot \varepsilon_3 \Rightarrow \varepsilon_3 = \frac{E_{ct}}{E_m} \varepsilon_c \,. \tag{A10}$$

From Eqs. (A4), (A10), we have

$$\varepsilon_c \cdot c = \varepsilon_1 \cdot a + \frac{E_c}{E_m} \varepsilon_c \cdot s \Rightarrow \varepsilon_c \left[c - \frac{E_{ct}}{E_m} \cdot s \right] = \varepsilon_1 \cdot a \Rightarrow$$

$$\Rightarrow \varepsilon_c \cdot (E_m \cdot c - E_{ct} \cdot s) = \varepsilon_1 \cdot a \cdot E_m \Rightarrow \varepsilon_c = \frac{\varepsilon_1 \cdot a \cdot E_m}{E_m \cdot c - E_{ct} \cdot s} \,. \tag{A11}$$

From Eqs. (A9), (A11), (A3), we have

$$E_{ct} \cdot \frac{\varepsilon_1 \cdot a \cdot E_m}{E_m \cdot c - E_{ct} \cdot s} (b + k) \cdot c$$
$$= E_f \cdot \varepsilon_1 \cdot a \cdot b + E_m \cdot \varepsilon_1 \cdot [(b + k) \cdot c - a \cdot b] \Rightarrow$$
$$\Rightarrow E_{ct} \cdot [a \cdot E_m (b + k) \cdot c + s \cdot E_f \cdot a \cdot b$$
$$+ s \cdot E_m [(b + k) \cdot c - a \cdot b]]$$
$$= E_m \cdot [E_f \cdot a \cdot b \cdot c + E_m \cdot c \cdot [(b + k) \cdot c - a \cdot b]],$$

and the elastic modulus of the composite is obtained as:

$$E_{ct} = \frac{E_m \cdot [E_f \cdot a \cdot b \cdot c + E_m \cdot [(b+k) \cdot c - a \cdot b) \cdot c],}{E_f \cdot a \cdot b \cdot s + E_m \cdot [a \cdot c \cdot (b+k) + s \cdot [(b+k) \cdot c - a \cdot b]]} . \tag{A12}$$

The volume fraction of the inclusion is

$$u_f = \frac{a^2 \cdot b}{c^2(B+k)} . \tag{A13}$$

For $k = 0$ (continuous inclusion), Eq. (A13) becomes

$$u_f = \frac{a^2}{c^2} , \tag{A14}$$

and Eq. (A12) becomes:

$$E_{ct} = \frac{E_m \cdot [E_f \cdot a \cdot c + E_m \cdot (c^2 - a \cdot c)]}{E_f \cdot a \cdot s + E_m [a \cdot c + s \cdot c - a \cdot s]} . \tag{A15}$$

From the geometry of the model: $s = c - a$

Thus, Eq. (A15) becomes:

$$E_{ct} = \frac{E_m [E_f \cdot u_f^{1/2} + E_m (1 - u_f^{1/2})]}{E_f (u_f^{1/2} - u_f) + E_m [u_f - u_f^{1/2} + 1]} . \tag{A16}$$

For $k = s$ (end-to-end spacing of the inclusions is equal to side-to-side spacing), Eq. (A12) becomes:

$$E_{ct} = \frac{E_m [E_f \cdot a \cdot b \cdot c + E_m \cdot [(b+s) \cdot c - a \cdot b] \cdot c]}{E_f \cdot a \cdot b \cdot s + E_m [a \cdot c \cdot (b+s) + s[(b+s) \cdot c - a \cdot b]]} \tag{A17}$$

and the volume fraction of the inclusion

$$u_f = \frac{a^2 b}{c^2 (b+s)} \tag{A18}$$

General case

Now let us consider the general case where n inclusions are arranged in the direction of the tensile load and v inclusions in the perpendicular direction, as illustrated in Fig. 1b.

Under the action of a tensile load F_c acting perpendicularly to the long sides of the inclusion prisms, the model shown in Fig. (1b) may be considered to consist of a sequence of two parts. One of these parts consists of the inclusion prisms (1), with cross-sectional area $a \cdot b$ in parallel with a matrix prism (2). The other part consists of the remainder of the matrix material (3) in series with (1) + (2). In Fig. 1b, $s = l - na$ denotes the side-to-side spacing of the inclusions and k a parameter depending on the inclusion length and, which becomes zero when the inclusion is continuous.

Equilibrium of forces in the direction of the applied uniaxial tensile force F_c (or stress σ_c) gives the following equation where the subscripts $1 - 3$ refer to the different layers, or parts of layers of the model:

$$\sigma_c \cdot (b+k) \cdot c = \sigma_3 \cdot (b+k) \cdot c \Rightarrow \sigma_c = \sigma_3 \tag{A19}$$

$$\sigma_c \cdot (b+k) \cdot c = \sigma_1 \cdot v \cdot a \cdot b + \sigma_2 [(b+k) \cdot c - v \cdot a \cdot b] . \tag{A20}$$

If it is assumed that there is "perfect bonding" between the inclusion and the matrix then:

$$\varepsilon_1 = \varepsilon_2 . \tag{A21}$$

From the geometry of the model the total expansion is:

$$\varepsilon_c \cdot l = n \cdot \varepsilon_1 \cdot a + \varepsilon_3 (l - n \cdot a) . \tag{A22}$$

The constitutive equations, assuming proportionality of stress to strain are:

$$\varepsilon_c = E_{ct} \cdot \varepsilon_c \tag{A23}$$

$$\sigma_1 = E_f \cdot \varepsilon_1 \tag{A24}$$

$$\sigma_2 = E_m \cdot \varepsilon_2 \tag{A25}$$

$$\sigma_3 = E_m \cdot \varepsilon_3 , \tag{A26}$$

where the notation concerning the elastic moduli, the stresses, and the strain is the same as in the previous part. The combination of Eqs. (A19), (A23),(A26) gives

$$E_m \cdot \varepsilon_3 = E_{ct} \cdot \varepsilon_c \Rightarrow \varepsilon_3 = \frac{E_{ct}}{E_m} \cdot \varepsilon_c . \tag{A27}$$

From Eqs. (A22), (A27), we have

$$\varepsilon_c \cdot l = n \cdot \varepsilon_1 \cdot a + \frac{E_{ct}}{E_m} \varepsilon_c \cdot (l - n \cdot a)$$

$$\Rightarrow \varepsilon_c \left[l - \frac{E_{ct}}{E_m} (l - n \cdot a) \right] = n \cdot \varepsilon_1 \cdot a \Rightarrow$$

$$\Rightarrow \varepsilon_c = \frac{n \cdot \varepsilon_1 \cdot a \cdot E_m}{l \cdot E_m - E_{ct}(l - n \cdot a)} . \tag{A28}$$

From Eqs. (A20), (A21), (A23), (A24), (A25), (A28), we have:

$$E_{ct} = \frac{n \cdot \varepsilon_1 \cdot a \cdot E_m}{l \cdot E_m - E_{ct}(l - n \cdot a)} (b+k) \cdot c = E_f \cdot \varepsilon_1 \cdot v \cdot a \cdot b$$

$$+ E_m \cdot \varepsilon_1 [(b+k) \cdot c - v \cdot a \cdot b]$$

$$\Rightarrow E_c [n \cdot a \cdot E_m (b+k) \cdot c + (l - n \cdot a) \cdot E_f \cdot v \cdot ab$$

$$+ (l - n \cdot a) \cdot E_m [(b+k) \cdot c - v \cdot a \cdot b]]$$

$$= E_m [l \cdot E_f \cdot v \cdot a \cdot b + l \cdot E_m [(b+k) \cdot c - v \cdot a \cdot b]] \Rightarrow ,$$

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and the elastic modulus of the composite is obtained as:

$$\Rightarrow E_{ct} = \frac{E_m \cdot [E_f \cdot v \cdot a \cdot b \cdot l + l \cdot E_m[(b+k) \cdot c - v \cdot a \cdot b]]}{E_f \cdot v \cdot a \cdot b \cdot (l - n \cdot a) + E_m[n \cdot a \cdot (b+k) \cdot c + (l - n \cdot a)[(b+k) \cdot c - v \cdot a \cdot b]]} \,. \quad \text{(A29)}$$

The volume fraction of the inclusion is given as:

$$U_f = \frac{v \cdot n \cdot a^2 \cdot b}{c \cdot l \cdot (b+k)} \quad \text{(A30)}$$

For $k = 0$ (continuous inclusions)

$$U_f = \frac{v \cdot n \cdot a^2}{c \cdot l} \quad \text{(A31)}$$

and the elastic modulus of the composite becomes

$$E_{ct} = \frac{E_m \cdot [E_f \cdot v \cdot a \cdot l + E_m \cdot l \cdot [c - v \cdot a]]}{E_f \cdot v \cdot a(l - n \cdot a) + E_m \cdot [l \cdot c - v \cdot l \cdot a + n \cdot v \cdot a^2]} \quad \text{(A32)}$$

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