EVALUATION OF THE ELASTIC MODULUS OF PARTICULATE COMPOSITES BY MEANS OF A SPHERE-WITHIN-CUBE AND A CIRCLE-INTO-SQUARE MODELS AND THE EFFECT OF FILLER SHAPE.

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Abstract

In this paper the elastic modulus of particulate composites is evaluated, by means of two models, namely the sphere-within cube and the circle into square formations. The theoretical results predicted by the above representative volume elements are compared with formulae existing in the literature as well as with experimental results carried out through tension experiments and ultrasonic measurements in iron/epoxy particulate composites. The tensile experimental values of the elastic modulus (Ec) were found well below those of ultrasonic measurements. The theoretical values of Ec predicted by the first model are close to those of ultrasonic measurements for high filler volume fractions, while the respective ones predicted by the second model are close to those obtained from tensile experiments.

Introduction

The characterization of a composite system consisting of a matrix in which filler particles are dispersed has not been achieved to date; parameters as the size, the shape, aspect ratio and distribution of reinforcing particles affect the mechanical properties of a composite. An also important parameter is the adhesion quality between matrix and filler, as well as the interaction between fillers mainly for high filler volume fractions.

A number of theoretical analyses, which define the elastic modulus of particulate composites is resulted from theories of rigid inclusions in non rigid matrix, where the enhancement in elastic modulus is considered to be analogous to the increase in viscosity[1]. A greater number of theoretical analyses is derived, from theories of rigid inclusions in rigid matrix, where a representative volume element is necessary, whose properties are generalize for all the composite. [1,2] The analyses range from simple ones, where a kind of law of mixture is used, to more sophisticated methods including a self consistent model [3-5]variational [6] and exact [7,8] methods based on elasticity theory. In ref. [9] the influence of the interphace zone as a third phase on the elastic modulus values is considered, while in ref. [10] a concept of the interaction between fillers on the elastic modulus is reported.

In this paper the elastic modulus of particulate composites is evaluated. For this purpose a sphere-within-cube model and another one in circle-into-square formation are considered. The predicted values by the first model, for high filler content, are close to those predicted by ultrasonic measurements in epoxy/iron particulate composite, which are well above the tensile experimental results. The values derived from the second model approximate satisfactorily the tensile experimental results.

Theoretical Considerations

The theoretical analysis is based on the following assumptions.

1) The matrix and the inclusions are homogeneous and isotropic materials.
2) The strains are small enough to maintain linearity between stress-strain relations.
3) There is perfect adhesion between matrix and filer.

For the model presents in Fig(1), named model 1, the filler volume fraction is given

$$u_f = \frac{4\pi r^3}{3c^3} \qquad (1)$$

For the model presented in Fig (2), named model 2 the filler volume fraction is given by

$$u_f = \frac{\pi r^2}{c^2} \qquad (2)$$

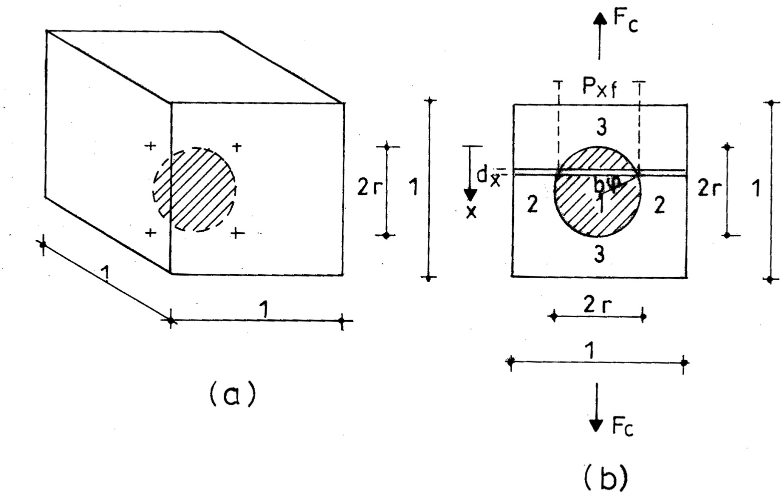

Fig. 1 (a): Sphere-within-cube model.
(b): Vertical section, in which the thickness dx, of an horizontal strip is shown.

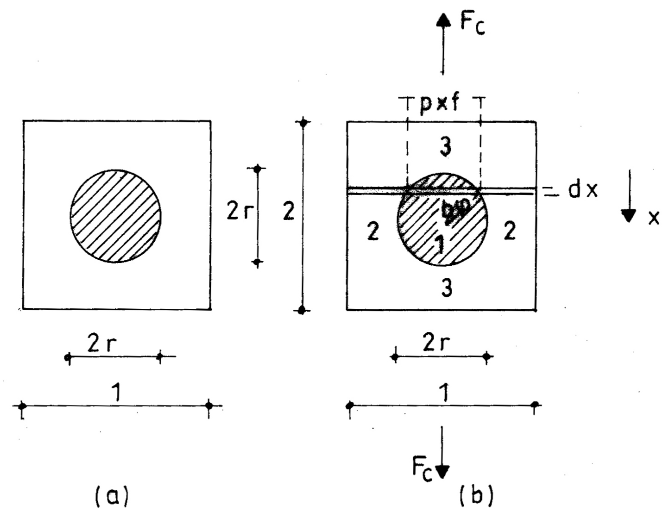

Fig. 2 (a): Circle-into-square model.
(b): parts formed in the model and the thickness dx of a strip formed by horizontal sections.

Model 1

The elastic modulus of a layer with thickness dx (fig (1b)) is given by

$$E_x = \frac{\pi\rho_{xf}^2 E_f + \left(c^2 - \pi\rho_{xf}^2\right)E_m}{c^2} \tag{3}$$

where the indices f and m, correspond to the filler and matrix respectively and ρ_{xf} is the radius of the circle of the laxer with thickness dx (fig (1b)).

The elastic modulus $E_{1,2}$, of components 1 (1) and (2) is obtained as follows

$$\frac{1}{E_{1,2}} = c\int_0^{\pi/2} \frac{\sin\varphi \, d\varphi}{E_m c^2 + \left(E_f - E_m\right) r^2 \sin^2\varphi} \tag{4}$$

where r is the radius of the spherical inclusions and φ is the angle shown in fig (1b).

After the integration one obtains the following expression for $E_{1,2}$.

$$E_{1,2} = \frac{2\pi r \cdot E_m (m-1)}{c^2} \cdot \sqrt{r^2 + \frac{c^2}{(m-1)\pi}} \cdot \frac{1}{\ell n \left[\dfrac{\sqrt{r^2 + \dfrac{c^2}{(m-1)\pi}} + r}{\sqrt{r^2 + \dfrac{c^2}{(m-1)\pi}} - r} \right]} \tag{5}$$

where $m = E_f / E_m$

The elastic modulus, E_c, of the composite is now obtained by considering the components (1,2) and (3) , which are in series, as follows

$$\mathrm{E}_c = \frac{\mathrm{E}_m \cdot \mathrm{E}_{1,2} \cdot c}{\mathrm{E}_{1,2}(c - 2r) + \mathrm{E}_m 2r} \tag{6}$$

Model 2

The elastic modulus of a layer with thickness dx (fig. (2b)) is given by

$$\mathrm{E}_\chi = \frac{2 p_{xf}\, \mathrm{E}_f + \left(c - 2 p_{\chi f}\right) \mathrm{E}_m}{c} \tag{7}$$

where $2 p_{xf}$ is the cord of the circle corresponding to the angle 2□, (fig. (2b)).

The elastic modulus $\mathrm{E}_{1,2}$ of the components (1) and (2) is obtained as follows

$$\frac{1}{\mathrm{E}_{1,2}} = \frac{c}{2}\int_{o}^{\pi} \frac{\sin\varphi \, d\varphi}{\mathrm{E}_m \cdot c + \left(\mathrm{E}_f - \mathrm{E}_m\right) r \sin\varphi} \qquad (8)$$

After the integration one finds the following equation for $\mathrm{E}_{1,2}$.

$$\frac{1}{\mathrm{E}_{1,2}} = \frac{c}{\mathrm{E}_m (m-1)\cdot r}\left[\frac{\pi}{2} - \frac{c}{\sqrt{(m-1)^2 c^2 - c^2}}\, x\ell n\left[\frac{(m-1)r + c + \sqrt{(m-1)^2 r^2 - c^2}}{(m-1)r + c - \sqrt{(m-1)^2 r^2 - c^2}}\right]\right] \qquad (9)$$

where $m = E_f / E_m$

The elastic modulus of the composite is obtained by considering the components (1,2) and (3) which are in series, and is now given by eqn (6).

Results and Discussion

By means of the present study except of the evaluation of the elastic modulus of particulate composites, the influence of the filler shape on the elastic modulus values through characteristic filler shapes is also examined. For this purpose the matrix volume distribution of each individual filler is maintained constant. Then the model sphere-within-cube is compared with the formation cube-within-cube, and the model circle-into-square is compared with the formation square-into-square. The appeared discrepancies among the above models are expected as the relationship between the radius r of the sphere and the side a of the cube of the inclusion is 2r=1,241□, while for the circle-into-square and square-into-square formations one finds 2r=1,129□, where r is the radius of the circle and □is the side of the square of the inclusion. Therefore for the force direction (figs (1b,2b) the matrix distance in this direction in models sphere-within-cube and circle-into-square is lower than of the formations cube-within-cube and square into square respectively. As a result and given that the elastic modulus of the filler is very high (60 times greater than that of the matrix) one finds lower deformations in the first two models and thus greater values for the elastic modulus than those of the respective second two models

In fig (3) the elastic modulus is plotted against filler volume fraction. The theoretical results for E_c predicted by the model sphere-within-cube are very high compared with the tensile experimental results especially for high filler content where they approximate the ultrasonic measurements. Although this model is realistic having physical meaning, however compared with the tensile experimental results does not seem to be formed in the composite.

However a doubt is mainted coorelated to the adhesion between matrix and filler due to the fact that current ages were not used in the specimens of the tensile experiments.

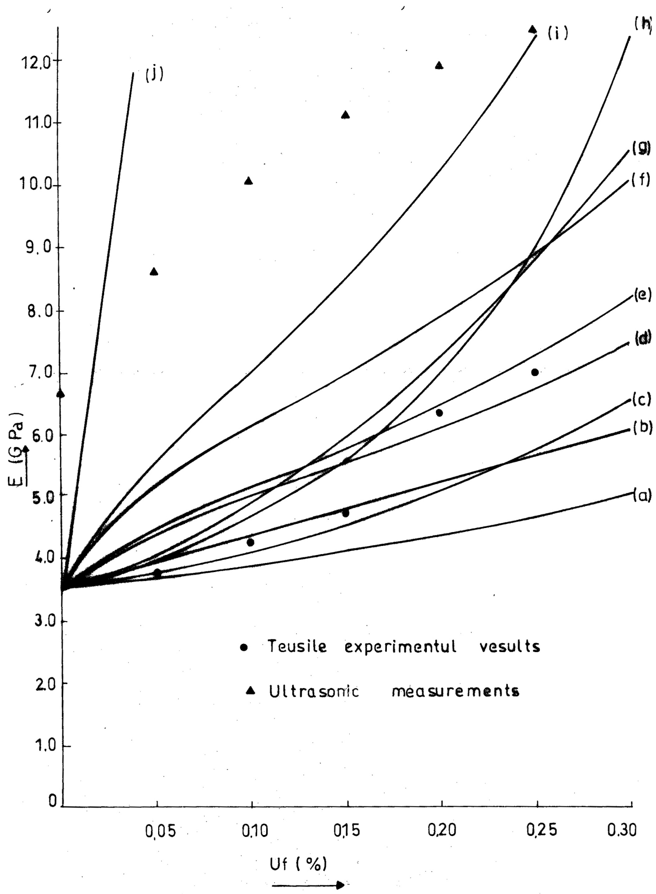

Fig. 3 Elastic modulus versus filler volume fraction in iron epoxy particulate composites. (a) ; eqn (A8), (b); eqn (A1), (c); eqn (A5), (d); eqn (A6), (e); circle into square model, (f); eqn (A4), (g); eqn (A2), (h); eqn (A3), (l); sphere-within-cube model, (j); eqn (A7).

Another part of the appeared discrepancies is probably due to the fact that the iron particles that were used in the specimens were not spherical in combination to the fact that a uniform distribution of the filler into the matrix volume did not exist. This model compared with the cube-within-cube formation gives for u_f=0,25, a value for E_c 39,1% greater than the later model, which gives 26,3% greater value than the experimental one. Here it is worth mentioning if a square prisma-within-cube model, where the axis of the prisma is in the force direction and has a dimension b=1,241, with the side of o cubical inclusion with the same volume, gives almost the same values as the model sphere-within cube. This last model presents an aspect ratio

$\lambda = \dfrac{1,241\alpha}{0,898\alpha} = 1,382$, where 0,898□is the basis side of the square-prisma.

In the same figure we observe that the predicted values for E_c as much by the model circle into square as by the model square-into-square, although both models are idealized ones for the particulate composite, approximate satisfactory by the experimental results. For high filler content the experimental results are bounded from the theoretical results predicted by the above models, where the appeared bounds are very small. In this case the appeared discrepancies are smaller than those of the previous models, as the difference between the diameter of the circle and the side of the square is small. Here also a model of rectangular-into-square with the side b of rectangular in the force direction b=1,129□, where □the side of the square, gives values for the E_c close to those predicted by the model circle into square.

In the same figure the curves predicted by the models Einstein [11], Guth-Smalwood [12,13], Mooney [14] and Ishay-Cohen [16]curves (b), (g), (h) and (c) respectively are traced too. These curves approximate the tensile experimental results for low filler content. As we can also observe from this figure, all the curves are bounded by those predicted by the low of mixture, curve (j) and the inverse low of mixtures curve (a).

The discrepancies anyhow between static and ultrasonic measurements can be attributed except the interpretation of ref [21] also to the fact that entanglements among the molecules which for static measurements do not influence the values of E_c, however probably can be detected by means of the ultrasonic method, where inertia phenomena increase the elastic modulus values.

Finally, in the diagram of fig, (4) it is observed that the values of the reinforcement coefficient by the ultrasonic method are very close to those derived from square into square and circle into square models.

The obtained high values for the elastic modulus of the matrix by means of the ultrasonic method are supported from the results of refs [22,23].

The tensile experimental values are derived from ref [19], while the ultrasonic measurements from ref [20].

The elastic constants of epoxy matrix and iron particles used in the specimens are the following

$$E_{m} = 3,5 GP\alpha \qquad E_f = 210 GP\alpha$$

$$V_{m} = 0,36 \qquad V_f = 0,29$$

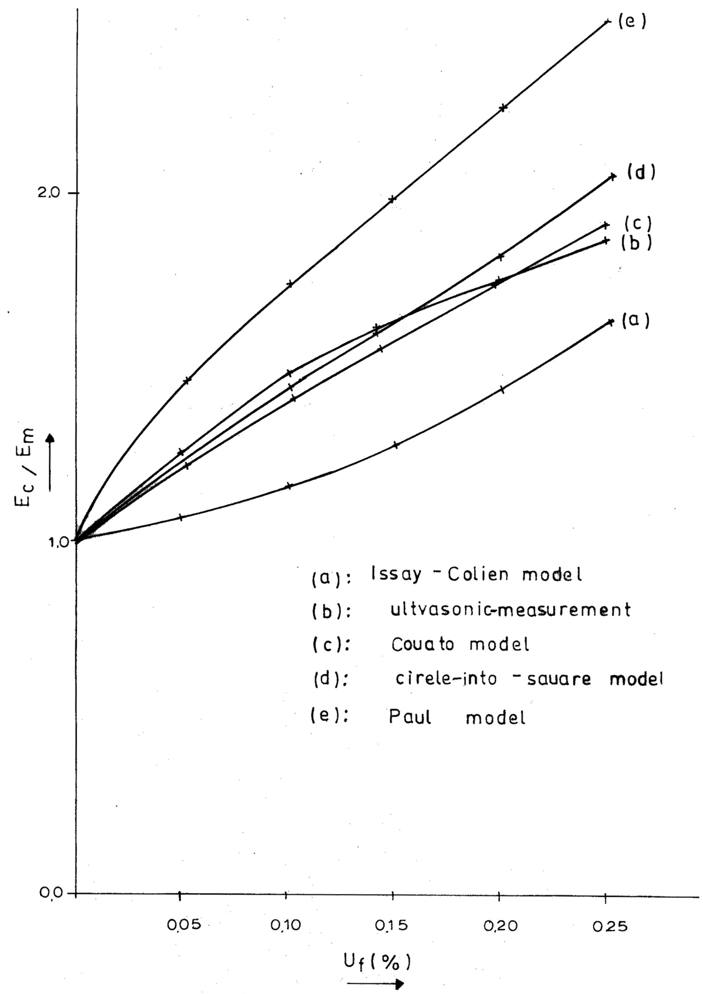

Fig. 4 Renforcement coefficients versus filler content in iron/epoxy particulate composites.

Conclusions

The theoretical results for E_c by means of the sphere within cube model present discrepancies from the tensile experimental results. For high filler content approximate the ultrasonic measurements.

The obtained values for E_c by means the circle into square model are close to tensile experimental results.

Differences in filler shape in the load direction of the order of 24,1% result to differences in E_c of the order of 39,1%, for $u_f = 0,25$.

The reinforcement coefficients obtained by ultrasonic measurements are close to those predicted by the circle-into-square and square-into-square models.

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Appendix

Equations existing in the literature and used for comparison are the followings.

1. Einstein formula [11]

$$E_c = E_m\left(1+2{,}5u_f\right) \qquad \text{(A1)}$$

2. Guth and Smallwood equation [12,13]

$$E_c = E_m\left(1+2{,}5u_f+14{,}1u_f^2\right) \qquad \text{(A2)}$$

3. Mooney equation [14]

$$E_c = \exp\frac{1+2{,}5u_f}{1-5\cdot u_f} \qquad \text{(A3)}$$

where s=1,35 for closed spheres package.

4. Paul model [15]

$$E_c = E_m\left(\frac{1+(m-1)u_f^{2/3}}{1+(m-1)\left(u_f^{2/3}-u_f\right)}\right) \qquad \text{(A4)}$$

where $m = E_f / E_m$

5. Ishay-Cohen model [16]

$$E_c = E_m\left(1+\frac{u_f}{\frac{m}{m-1}-u_f^{1/3}}\right) \qquad \text{(A5)}$$

where $m = E_f / E_m$

6. Counto equation [17]

$$E_c = E_m \left(1 + \frac{u_f}{\frac{1}{m-1} + u_f^{1/2} - u_f} \right) \quad \text{(A6)}$$

7. Law of mixtures [18]

$$E_c = E_f u_f + E_m u_m \quad \text{(A7)}$$

where $u_f + u_m = 1$

8. Inverse law of mixtures [18]

$$\frac{1}{E_c} = \frac{u_f}{E_f} + \frac{u_m}{E_m} \quad \text{(A8)}$$

where $u_f + u_m = 1$