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MODEL FOR PREDICTION OF THE ELASTIC RESPONSE OF COMPOSITE
MATERIALS OVER WIDE RANGES OF CONCENTRATION

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MODEL FOR PREDICTION OF THE ELASTIC RESPONSE
OF COMPOSITE MATERIALS OVER WIDE RANGES OF CONCENTRATION

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ABSTRACT

A model for the prediction of the elastic response of composite materials over wide ranges of concentration is presented. The method is based on the mathematical analogy between the motion of particles suspended in viscous media and the elastic deformation of solids. The system is locally described by linearized forms of the elastic moduli. The validity of these relations is then extended to all concentrations under the assumption that any new portion of filler "sees" the existing composite as a non-interacting homogeneous matrix. The method predicts the behavior of composites with solid spherical inclusions and foams over wide ranges of concentration. The model, free from adjustable parameters, shows excellent agreement with existing experimental data. The extension of the method to other inclusion geometries is straightforward.

INTRODUCTION

In the search of a mathematical model for the mechanical behavior of heterogeneous systems, a complete analogy is found between the basic equations for elasticity theory and those for the motion of suspended particles in viscous media. The analogy can be easily proved by writing the equations of motion for an incompressible material in terms of displacements:

$$-p_{,i} + Gu_{i,jj} + F_i = \rho \frac{\partial^2 u_i}{\partial t^2} \quad (1)$$

where G is the shear modulus, u_i is the displacement, F_i is the body force, p is the reactive pressure, and ρ is the density.

For comparison, the Navier-Stokes equations for an incompressible Newtonian fluid are:

$$-p_{,i} + n v_{i,jj} + \rho F_i = \rho \left(\frac{\partial v_i}{\partial t} + v_j v_{i,j} \right) \quad (2)$$

where n is the viscosity, v the velocity, and F the body force per unit mass. Equations 1 and 2 have a term by term equivalence, with the exception of the nonlinear component $v_j v_{i,j}$ representing a convective acceleration. Under creeping flow conditions, acceleration terms become irrelevant and a complete analogy can then be established. Given a certain geometry, the theoretical equations for a viscous flow problem and those for the elastic deformation of a solid are of the same form:

shear rate : viscosity

shear strain : shear modulus

For a suspension of rigid particles in an incompressible matrix, there is a simple relationship between relative viscosities and relative shear moduli

(relative to the unfilled matrix):

$$\frac{\eta}{\eta_m} = \frac{G}{G_m}$$

Thus, if a theory for the viscosity of a filled system or suspension is available, it can be used to estimate the shear modulus of an elastic system.

A second analogy can be also found in relation to the methods used in the solution of both problems, as indicated in Figures 1 and 2. In very general terms, it is found that there is an important group of contributions based on the establishment of an idealized geometry and packing arrangement (top of Figures 1 and 2). A second group is based on the determination of bounds, which avoids (or reduces) the idealization problem by use of energy and variational techniques (bottom of Figures 1 and 2). Typical limitations of geometric models and bound methods gave origin to a third group of contributions containing elements of both techniques (middle of Figures 1 and 2).

With regard to the problem of predicting the elastic performance of composite materials, (Figure 2), most of the models deal with specific idealized geometries and packing arrangements. These theories provide relations between any two independent elastic constants among a shear modulus (G), a bulk modulus (K) and a Young's modulus (E):

$$K = K(G_m, K_m, G_i, K_i, c)$$

$$G = G(G_m, K_m, G_i, K_i, c)$$

where subindices m and i refer to the matrix and the inclusion, and c is the volume fraction of the inclusion. Smith [11] noted that through algebraic

manipulation, most of these theories can be displayed in the form of a generalized "rule of mixtures":

$$G = G_m + (G_i - G_m)\phi = (1 - \phi)G_m + \phi G_i$$

where ϕ takes different functional forms. According to the most familiar contributions,

$$\phi = \frac{(8 - 10\nu_m)G_m + (7 - 5\nu_m)G_m}{(8 - 10\nu_m)G_i + (7 - 5\nu_m)G_m} c$$

(approximate Van der Poel) [11,16]

$$\phi = \frac{(8 - 10\nu_m)G + (7 - 5\nu_m)G_m}{(8 - 10\nu_m)G_i + (7 - 5\nu_m)G_m} c$$

(Kerner)[17]

$$\phi = \frac{(8 - 10\nu)G + (7 - 5\nu)G}{(8 - 10\nu)G_i + (7 - 5\nu)G} c$$

(Budiansky)[18]

where G and ν are the shear modulus and Poisson's ratio of the composite.

All of the previous models developed by elastic analysis, i.e., deformation of the matrix and inclusion, have a common feature: a specific idealized geometry and packing arrangement has to be assumed. In many composite materials, this regularity in geometry does not exist, and the material cannot be identified with any particular arrangement or ensemble valid throughout the whole structure. Thus, the previous treatments, in the strictest sense, do not truly represent a real composite. One way to avoid this idealization problem is

through the use of variational techniques, which do not need to use a precise geometric model.

The most elementary type of bounds can be calculated through the formalism of the complementary energy and strain energy theorems (Voigt [12], Paul [13]). However, the resulting bounds are often too far apart to be useful.

In order to improve these bounds, Hashin and Shtrikman [14] formulated new variational theorems on the basis of polarization tensors. However, limitations in the applicability of bounds still persist. In general, it is not known if the calculated bounds are indeed the most restrictive, and the problem becomes a trade-off between the amount of information introduced in the formulation, and the resulting contiguity of the bounds.

As a rule, it may be said that the usefulness of bounds is restricted to systems where the properties of the phases are similar, i.e., metal alloys. In polymer reinforcement, however, the differences between matrix and inclusion may be considerable. As a consequence, the upper and lower bounds tend to separate, making this technique inadequate for any predictive purpose.

PROCEDURE

As indicated earlier, a basic problem in the theory of viscosity of suspensions is the determination of correlations useful at high concentrations. Most of the contributions are based on extensions of Einstein's law of viscosity at infinite dilution.

The analogous problem of correlating the elastic properties of heterogeneous systems over wide ranges of concentration has not been successfully solved. As

will be discussed later, predictions according to different theories show a considerable lack of agreement.

The proposed model is based on a differential scheme similar to that used by Brinkman [3] and Roscoe [2] for the prediction of the viscosity of suspensions at high concentrations. In a later publication, Roscoe [9] tried to extend the method to the prediction of elastic properties of composites. McLaughlin [30] presented a similar differential scheme and evaluated its estimates in relation to the Hashin-Shtrikman bounds.

The present model is based on the calculation by elastic analysis of any two independent parameters for the composite, namely the shear modulus, the bulk modulus, or the Young's modulus. Calculations may proceed as follows [29]:

- 1) describe the deformation state (simple shear, etc.)
- 2) choose adequate coordinates for a given geometry,
- 3) state the deformation equations for matrix and inclusion,
- 4) state continuity of stress at the interface,
- 5) reduce volume integrals to surface integrals by Eshelby's formula [5],
- 6) integrate.

For spherical inclusions and dilute conditions, the shear modulus results as:

$$\frac{G}{G_m} = 1 - \frac{15(1 - \nu_m) [1 - (G_i/G_m)]}{2 - 5\nu_m + 2(4 - 5\nu_m)G_i/G_m} c = 1 + fc \quad (3)$$

and the bulk modulus is:

$$\frac{K}{K_m} = 1 + \frac{(1/K_m)(K_i - K_m)}{1 + [(K_i - K_m)/(K_m + (4/3)G_m)]} c = 1 + gc \quad (4)$$

where c notes the volume fraction, and the subindices m and i refer to the matrix and the inclusion.

For an infinitely rigid inclusion ($G_i \rightarrow \infty$) in an incompressible matrix ($\nu_m = 0.5$), equation (3) results in a relation similar to Einstein's law of viscosity:

$$\frac{\eta}{\eta_m} = \frac{G}{G_m} = 1 + 2.5 c \quad (5)$$

Consider a suspension containing a small concentration (volume) c_1 of spheres into which is placed a small concentration c_2 of spheres, and into this again a small concentration c_3 of spheres, and so on, up to n sets of spheres, and let the total concentration be c . Then on adding another set of spheres of concentration (volume fraction) c_{n+1} , the new total concentration (volume fraction) is:

$$c(1 - c_{n+1}) + c_{n+1} \quad (6)$$

So the increase in concentration is:

$$\Delta c = c_{\text{final}} - c_{\text{initial}} = c_{n+1}(1 - c) \quad (7)$$

If the original suspension is considered a homogeneous medium (matrix) around the new spheres which are in small concentrations, it is possible to express

the increment in shear modulus according to equation (3) as:

$$\frac{G_{n+1}}{G_n} = 1 + f c_{n+1} \quad (8)$$

Defining

$$\Delta G = G_{n+1} - G_n$$

and replacing c_{n+1} according to equation (7),

$$\frac{\Delta G}{G} = f c_{n+1} = f \frac{\Delta c}{1-c}$$

For infinitely small increments in concentration,

$$\Delta c \rightarrow dc$$

$$\Delta G \rightarrow dG$$

and

$$\frac{dG}{dc} = \frac{Gf}{(1-c)} \quad (9)$$

This differential expression for the increment in concentration can now be used in equation (3). After following a similar reasoning for the bulk modulus K , equations (3) and (4) become:

$$\frac{dG}{dc} = \frac{G \cdot 15(1-\nu) [1 - G_j/G]}{[7-5\nu + 2(4-5\nu)G_j/G](1-c)} \quad (10)$$

$$\frac{dK}{dc} = \frac{(K_j - K)}{1 + [(K_j - K)/(K + (4/3)G)](1-c)} \quad (11)$$

where $\nu = \frac{(3K - 2G)}{2(3K + G)}$ is the Poisson's ratio.

Equations (10) and (11) constitute a coupled system to be numerically integrated with the following boundary conditions:

$$\begin{array}{lll} c = 0 & G = G_m & \text{(matrix properties)} \\ & K = K_m & \\ & & (12) \\ c = 1 & G = G_i & \text{(inclusion properties)} \\ & K = K_i & \end{array}$$

RESULTS

1) Solid Inclusions

Smith [11] compared the behavior of well known models for the prediction of the mechanical response of composites, i.e., Van der Poel [16], Kerner [17], Budiansky [18], etc. In order to test the performance of the present model, the same parameter values were used in the simulation. Data in Figure 3a have been calculated with a shear modulus ratio $G_i/G_m = 30$ and Poisson's ratios $\nu_i = 0.25$ and $\nu_m = 0.40$, representing the case of glass spheres embedded in an epoxy matrix.

Figure 3b has been calculated with $G_i/G_m = 70,000$, $\nu_i = 0.25$, $\nu_m = 0.5$, representing the expected shear properties of glass spheres imbedded in a lightly vulcanized matrix of natural rubber.

In both cases, a considerable discrepancy among the different models is observed. The inconveniency of bounds calculated according to Hashin and Shtrikman [14] is also stressed. The predictions of the present simulation are close to those of Van der Poel [16].

Richard [7] presented experimental data for elastic modulus and Poisson's ratio of a polyester matrix reinforced with glass spheres ($G_m = 5.94 \cdot 10^8$ Pa, $\nu_m = 0.45$, $G_i = 2.90 \cdot 10^{10}$ Pa, $\nu_i = 0.21$). This author compared his experiments with the predictions of several theories. Figure 4 indicates the excellent prediction of the present model, while theories by Kerner [17] and Hill [20,21] only perform well at filler concentrations below 20 percent volume.

Richard's data [7] were also used by Smith [11] for comparison with other theoretical contributions as indicated in Figure 5a for relative moduli, and Figure 5b for Poisson's ratio. Calculations were performed with $G_m = 5.84 \cdot 10^8$ Pa, $\nu_m = 0.44$, $G_i = 2.90 \cdot 10^{10}$ and $\nu_i = 0.21$.

Smith [23] compared his own experimental findings on epoxy resin-glass spheres systems with several theoretical predictions. The properties of matrix and inclusion are $E_m = 2.68 \cdot 10^9$ Pa, $\nu_m = 0.394$, $E_i = 7.6 \cdot 10^{10}$ Pa, $\nu_i = 0.23$. Results from those predictions are plotted and compared with the present model in Figure 6.

4. Foams

Equations (10) and (11) have been integrated for the case of a foam by assuming negligible moduli for the inclusion,

$$G_i \rightarrow 0$$

$$K_i \rightarrow 0$$

The simulation was compared with the experimental results of Gent and Thomas [24] for a vulcanized rubber foam ($E_m = 2.59 \cdot 10^6$ Pa, $\nu_m = 0.49$). Figure 7 shows excellent agreement in the prediction of relative modulus versus relative density δ ,

$$\delta = \frac{\delta_{\text{foam}}}{\delta_{\text{rubber}}} = 1 - c \quad (13)$$

where c is, as before, the volume fraction of the inclusion.

The performance of the model in the prediction of foam properties was also tested with the experimental work of Moore et al. [25] for a variety of foamed thermoplastics (PVC, polypropylene copolymer, styrene-acrylonitrile copolymer, etc.). These authors observed that plots of relative modulus (relative to the solid matrix) versus relative densities (δ) could be correlated with a single curve in shear, tension and compression according to:

$$\frac{E_{\text{foam}}}{E_{\text{solid}}} = \left[\frac{\text{density of foam}}{\text{density of solid}} \right]^2 = (\delta)^2 = (1 - c)^2 \quad (14)$$

Figure 8 compares the predictions of several theories with the empirical findings of Moore et al. [25] and the results of the present model. Simulations were performed for a polypropylene copolymer characterized by $E_m = 1.13$ GPa, $\nu_m = 0.41$. Again, the proposed method shows total agreement with the experimental values.

3) Low modulus incompressible inclusions (rubbers)

For the limiting case of an incompressible matrix ($\nu_m = 0.5$) with $G_i = 0$,

equation (10) admits an analytical solution as

$$\frac{G}{G_m} = (1 - c)^{5/3} \quad (15)$$

which represents the behavior of rubber modified polymers.

Holliday et al. [31] compared several relevant theories for a case representing soft spheres in a rigid matrix (Figure 9). The experimental data correspond to acrylonitrile-butadiene-styrene (ABS) polymers which consist of a dispersion of soft polybutadiene rubber spheres in a styrene-acrylonitrile (SAN) copolymer matrix which has a relatively high modulus.

The different theories examined are not in good agreement. The proposed model was tested with typical values for styrene-acrylonitrile copolymer (SAN) ($E_m = 3.1 \cdot 10^{-3}$ MPa, $\nu_m = 0.38$) [32], and $E_i/E_m = 0$. Results presented in Figure 9 indicate good prediction of the experiments.

CONCLUSIONS

Despite many contributions, the problem of predicting the elastic behavior of composite materials over wide ranges of concentration has not been satisfactorily solved.

Different theories based on well defined geometric models present a considerable divergence of results. The use of bound methods has been proven inadequate for problems in polymer reinforcement, where solutions become a trade-off between the contiguity of the bounds and the amount of information (complexity) introduced in the model.

In the present work, a correct linearized relation for the elastic deformation of matrix and inclusion is assumed at each concentration. This relation, which is valid locally, can then be extended to all concentrations under the assumption that any new portion of filler "sees" the existing composite as a non-interacting homogeneous matrix.

The model, presenting no adjustable parameters, accurately represents available experimental data on solid spherical inclusions and foams. The method can be readily extended to other geometries for the inclusion.

ACKNOWLEDGEMENT

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FIGURE CAPTIONS

- Figure 1 Models for viscosity of suspensions.
- Figure 2 Models for prediction of elastic response in composite materials.
- Figure 3a Relative shear modulus versus volume fraction of filler for $G_i/G_m = 30$, $v_i = 0.25$, $v_m = 0.4$ (from Smith [11]). Solid curves, model performance; dashed curves, predictions with different models: (1), approximate Van der Poel [16]; (2), Kerner or Hashin and Shtrikman highest lower bound [14]; (3), corrected Van der Poel [16]; (4), Budiansky [18].
- Figure 3b Relative shear modulus versus volume fraction of filler for $G_i/G_m = 70,000$, $v_i = 0.25$, $m = 0.5$ from Smith [11]. Solid curve, model performance; dashed curves, predictions with different models. (1), approximate Van der Poel [16]; (2), Hashin and Shtrikman highest lower bound [14]; (3), corrected Van der Poel [16]; (4), Budiansky [18].
- Figure 4 Elastic modulus versus volume fraction of filler for the case of glass beads in a polyester matrix. Experimental data from Richard [19]. Solid curve, model performance; dashed curves, predictions according to 1), isostrain; 2), isostress; 3), Kerner [17]; 4), Hill [20,21].
- Figure 5 Relative Young's modulus E/E_m (5a) and Poisson's ratio (5b) versus volume fraction of filler for the case of glass beads in a polyester matrix. Experimental data from Richard [19]. Solid lines, model performance; dashed lines, predictions with different models. 1), Hashin and Shtrikman highest lower bound [14]; 2), Hashin and Shtrikman least upper bound [14]; 3), Van der Poel [16]; 4), Budiansky [18].

- Figure 6 Relative Young's modulus E/E_m versus volume fraction of filler for the case of glass spheres in an epoxy matrix. Experimental data by Smith [23]. Solid curve, model performance; dashed curves, predictions with models by 1), Hashin and Shtrikman highest lower bound [14]; 2), Hashin and Shtrikman least upper bound [14]; 3), Van der Poel [16]; 4), Budiansky [18].
- Figure 7 Young's modulus of a highly vulcanized rubber foam E_f relative to the solid rubber E_m versus volume fraction of rubber in the foam. Solid line, model performance. Experimental points were determined by Gent et al. [24].
- Figure 8 Relative moduli of foams versus relative densities. Calculations here have been performed for the thermoplastics described by Moore et al. [25]. Solid curves, model performance for 1), Young's modulus; 2), shear modulus. Dashed curves, predictions according to different models. 3), Kerner [17]; 4), square-in-square model [27]; 5), cube-in-cube model [27]; 6), law of mixtures. Moore et al. [25] correlated experimental moduli with a square law which results coincident with curve 1 (model prediction).
- Figure 9 Relative modulus of acrylonitrile-butadiene-styrene (ABS) copolymer versus volume fraction of polybutadiene. Experimental data by Holliday et al. [31]. Solid curve, model performance. Dashed curves, predictions with different models. 1), Van der Poel [16]; 2) Parallel; 3), Kerner [17]; 4), Reiner-Hashin [33]; 5), Series.

VISCOSITY OF SUSPENSIONS

GEOMETRIC
MODEL
SPECIFIED

LINEARIZED
SOLUTIONS AT
 ∞ DILUTION
EINSTEIN [1]

EXTENSION TO
HIGH CONC.
MOONEY [5]
ROSCOE [2]
BRINKMAN [3]
FARRIS [4]

ADDITIONAL INFORMATION ON
RANDOM GEOMETRY OF
SUSPENSION, PARTICLE
SHAPE, ETC.

WEISSBERG [6]

DETERMINATION
OF BOUNDS

MINIMUM
ENTROPY
PRODUCTION

HASHIN [7]
PRAGER [8]

ELASTIC ANALYSIS OF COMPOSITE MATERIALS

GEOMETRIC
MODEL
SPECIFIED

LINEARIZED
SOLUTIONS AT
 ∞ DILUTION
DEWEY [10]

EXTENSION TO
HIGH CONC.
KERNER [17]
VAN DER POEL [16]
BUDIANSKY [18]
ROSCOE [9]

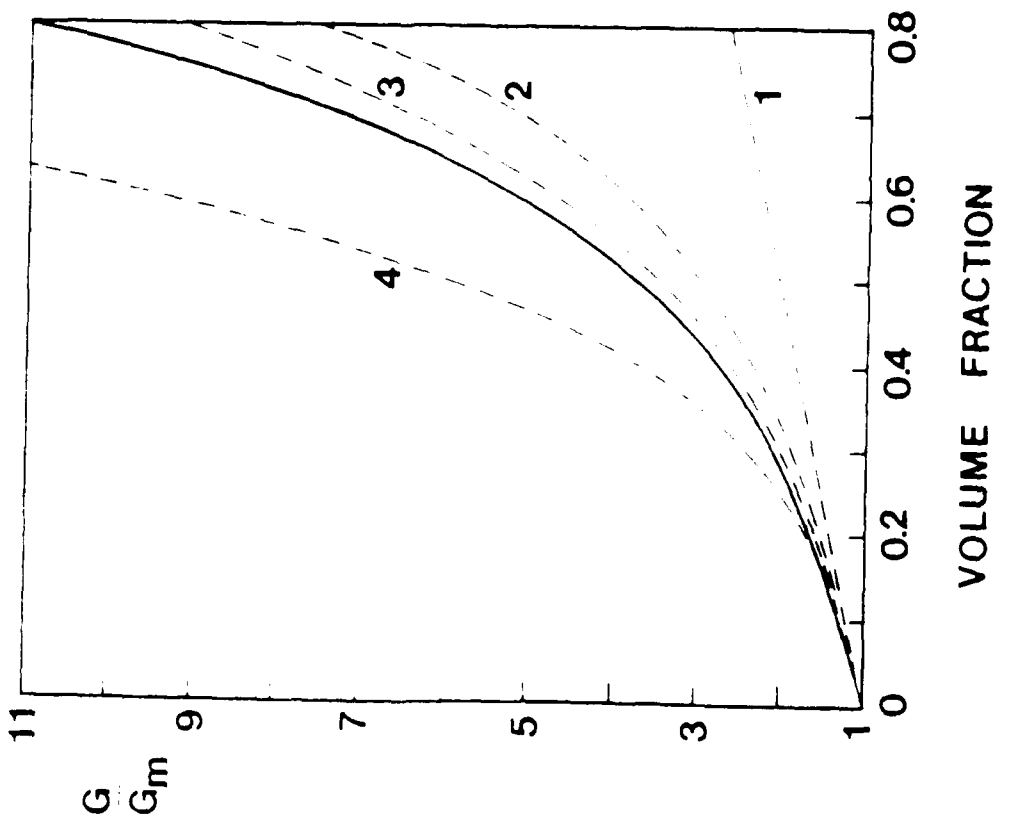
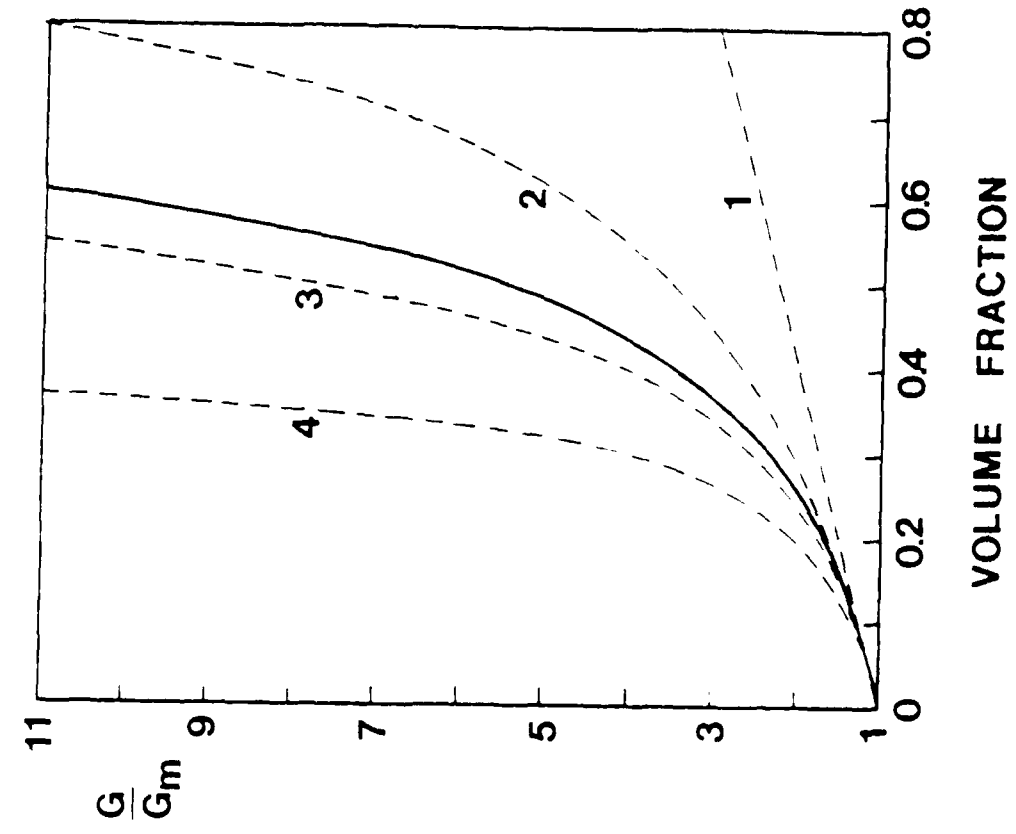
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ON PACKING
ARRANGEMENTS

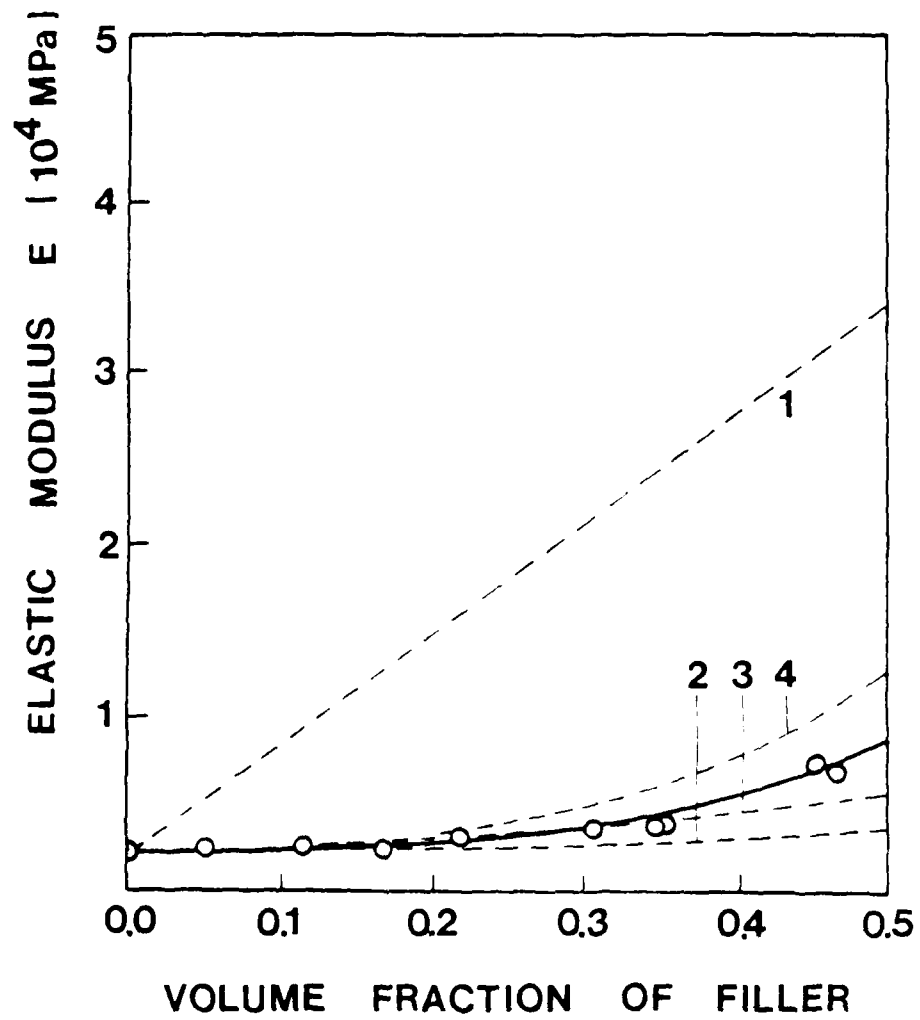
HASHIN [28]

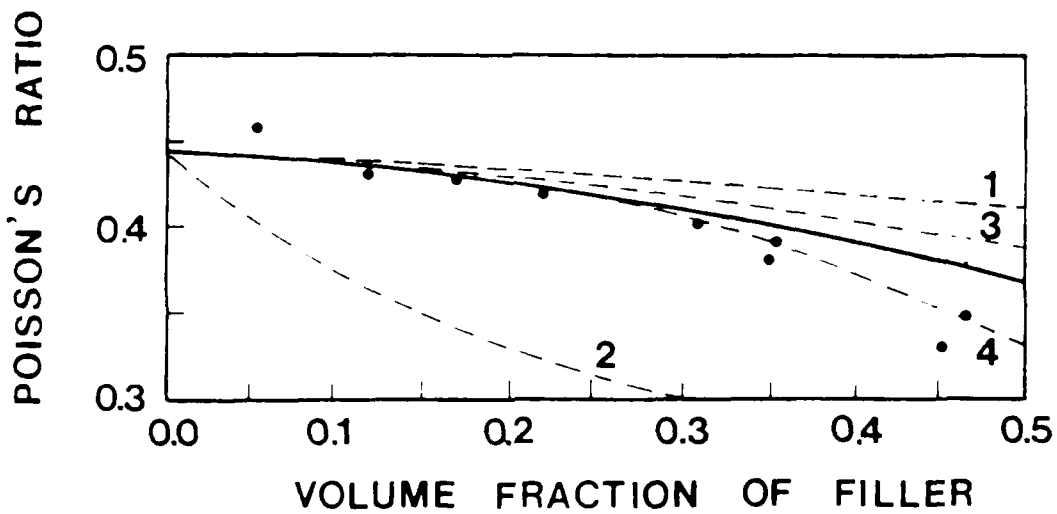
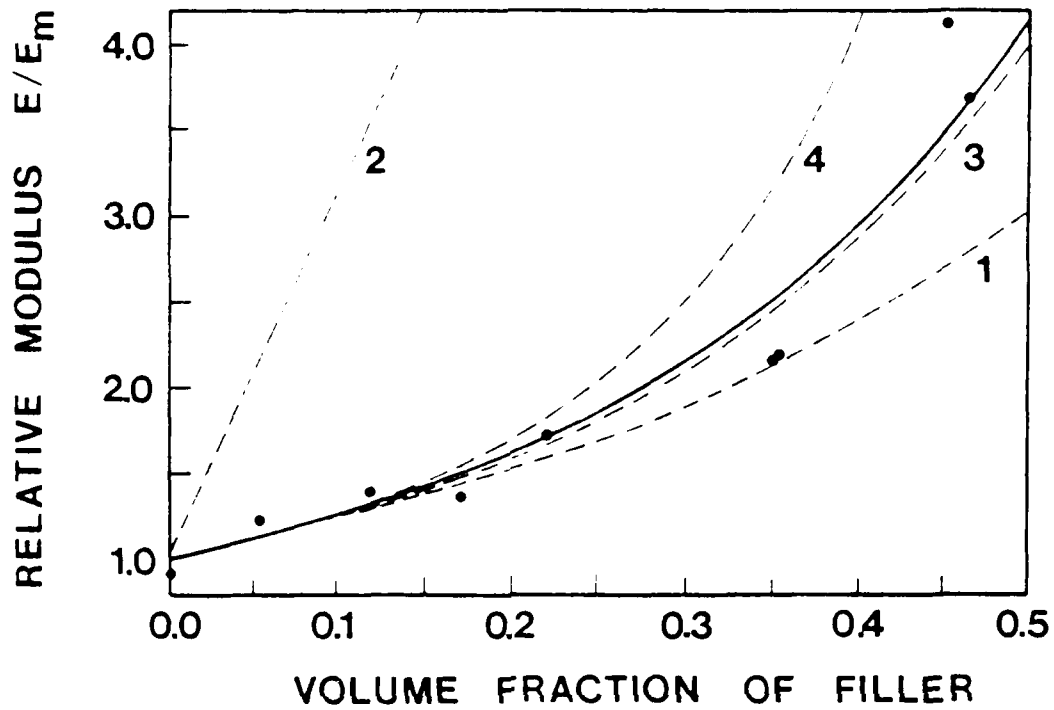
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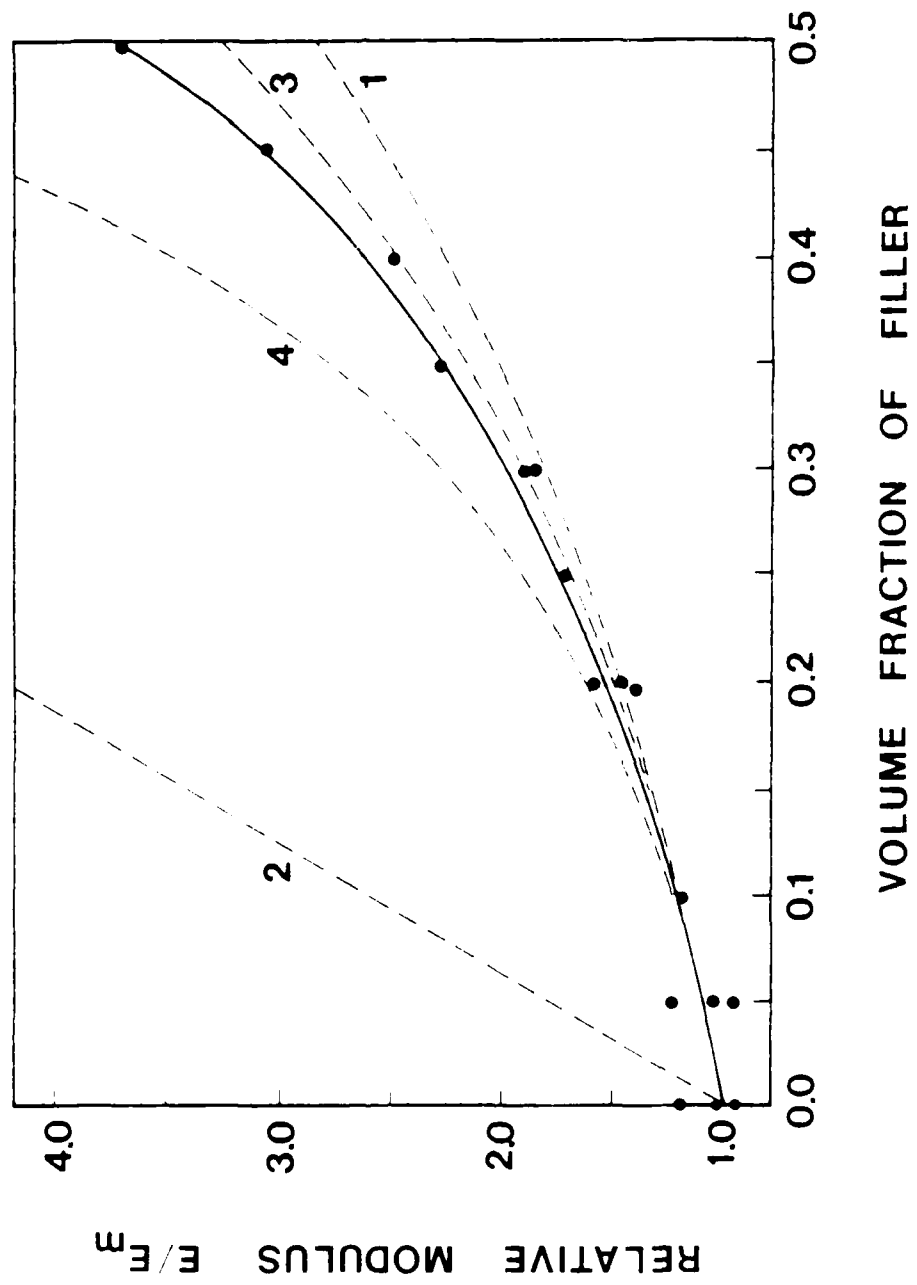
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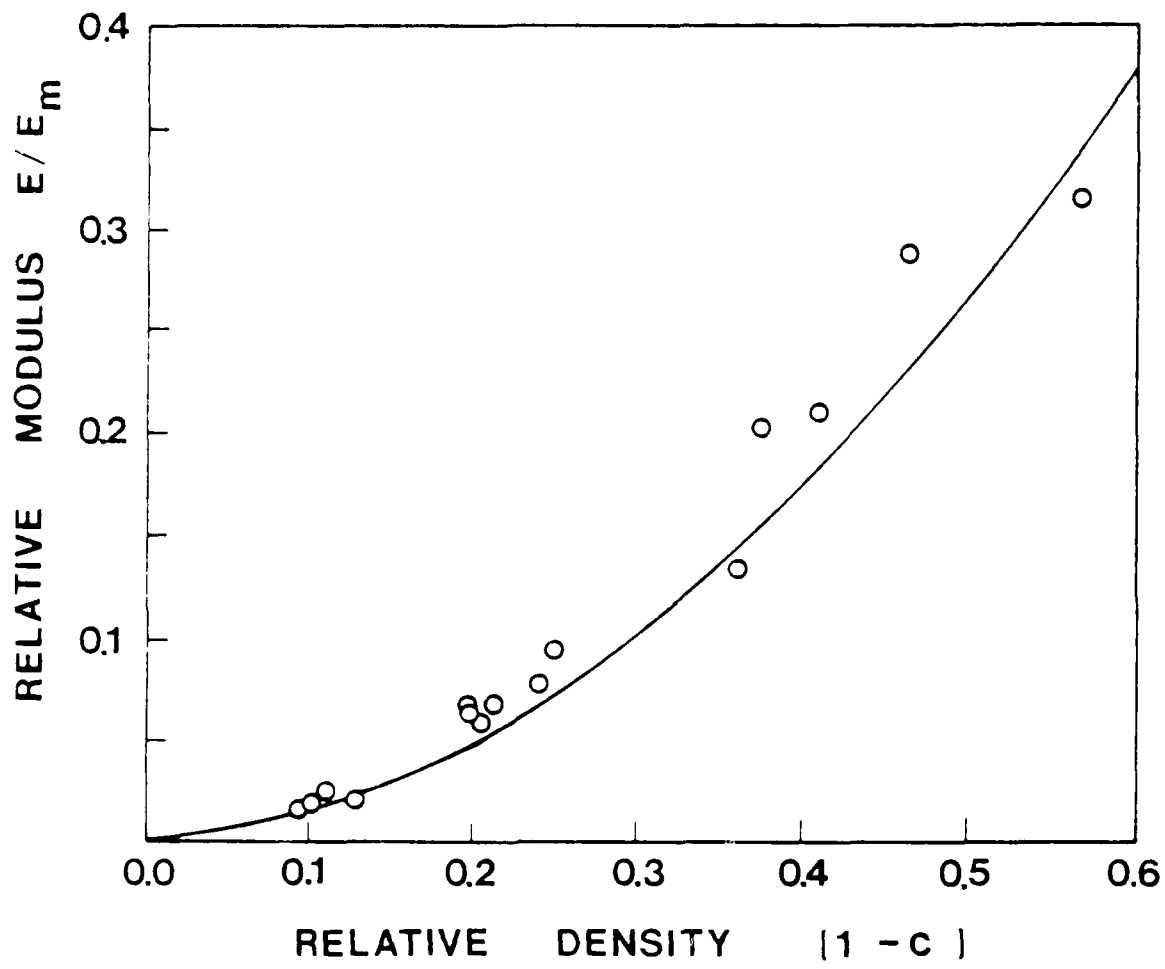
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PAUL [13]
HASHIN ET AL. [14]

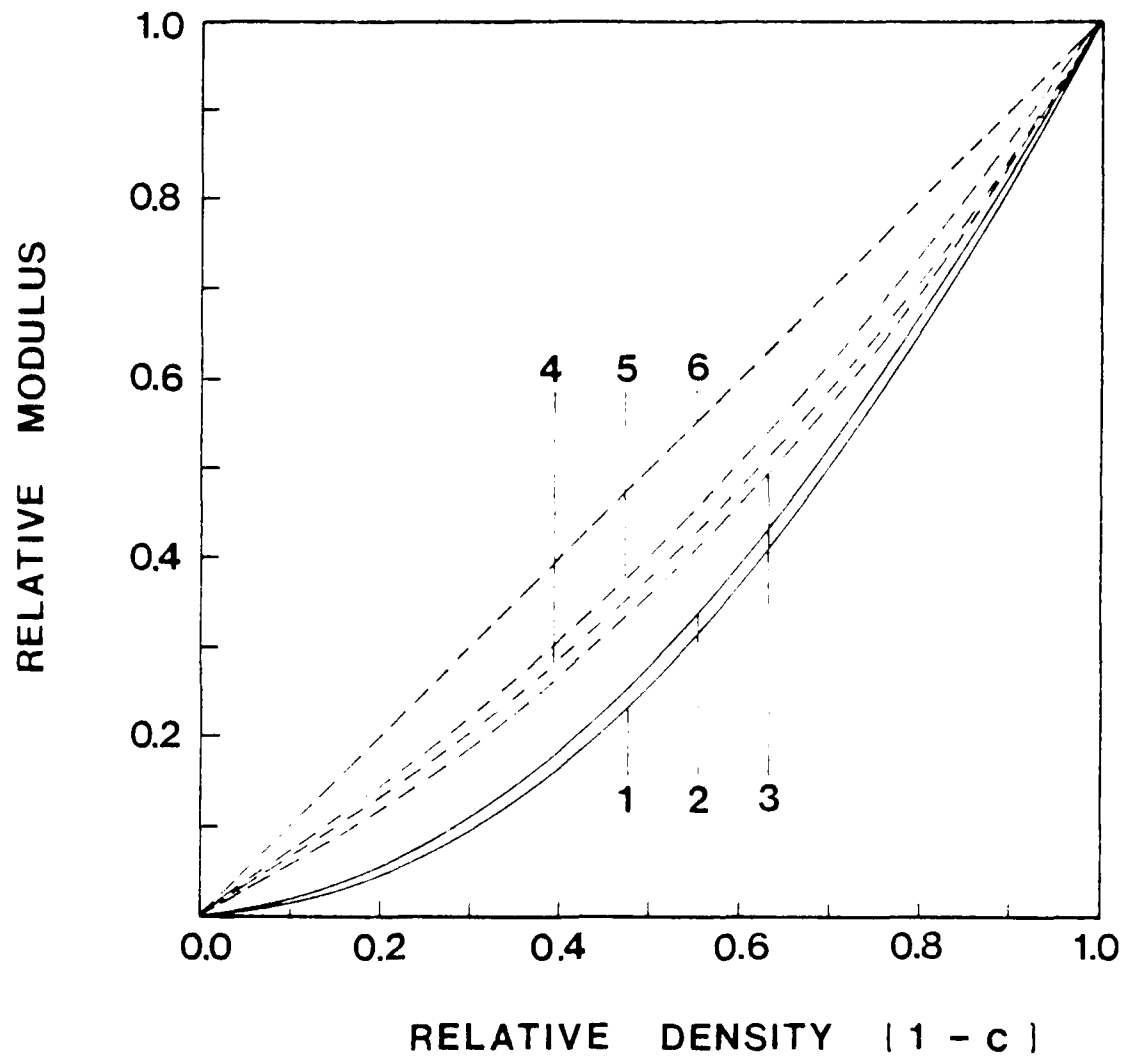


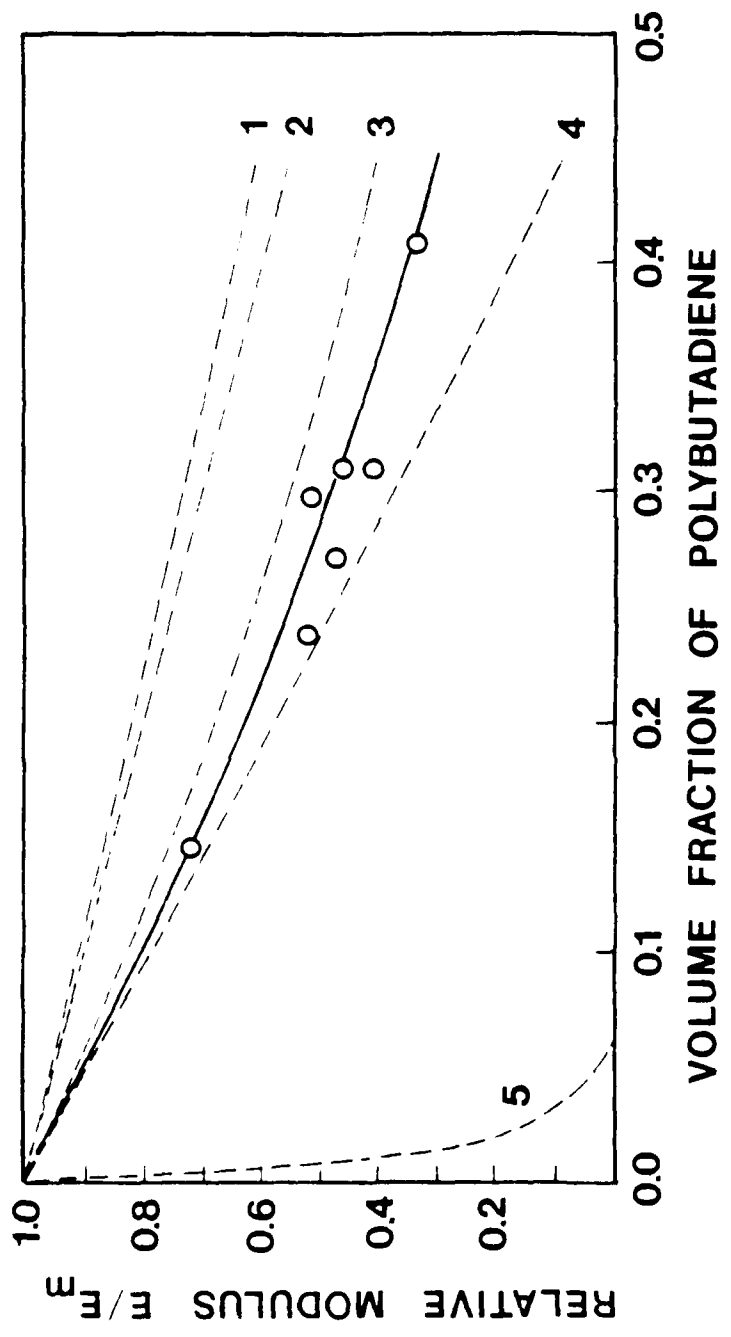












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