Polymer Vol. 37 No. 3, pp. 527–530, 1996

0032-3861/96/$15.00 + 0.00

A note on the Young's modulus of isotropic two-component materials*

A. J. Owen† and I. Koller
Department of Physics, University of Regensburg, 93040 Regensburg, Germany
(Received 18 May 1995)

A model is described for predicting the elastic behaviour of two-phase isotropic materials, based on Takayanagi's model, but taking transverse constraints into account. The model is compared with experimental results on SiO_2-filled epoxy and starch-filled poly-3-hydroxybutyrate (PHB) and PHB/3-hydroxyvalerate composites.

(Keywords: Young's modulus; two-component materials; transverse constraint)

Introduction

Predicting the elastic moduli of two-component materials from the properties of the individual components has been the subject of many investigations[1–11]. There is no exact general solution to this elasticity problem, since the moduli are affected by the morphology of the system, i.e. on the juxtaposition and shape of the individual components in space, and how they are bonded together. The basic difficulty is that one does not know *a priori* how stress and strain are transmitted through the system.

Takayanagi *et al.*[10] proposed 'series' and 'parallel' models for polymers containing two separate phases. In their series model, the Young's modulus E is obtained by averaging the strains in the individual phases, giving:

$$E^{-1} = (1-\phi)E_M^{-1} + \phi E_F^{-1} \tag{1}$$

where E_M and E_F are the Young's moduli of the two components and ϕ is the volume fraction of the component labelled F. Similarly, by averaging stresses, the parallel model gives:

$$E = (1-\phi)E_M + \phi E_F \tag{2}$$

It has been discussed by Arridge[12] that E in equation (1) is a lower bound, which is related to the well known Reuss bounds for shear and bulk moduli[13]. Equation (2), known as the 'rule of mixtures', is related to the upper bounds for shear and bulk moduli due to Voigt[14], but is not identical to them.

Since neither of these bounds generally describes the behaviour of a two-phase composite, Takayanagi developed a combined series–parallel model for the tensile modulus E, by introducing a degree of parallelinity into the series model[15].

One obvious shortcoming of Takayanagi's model is that it does not take into account transverse constraints on the components due to Poisson contraction, i.e. it is essentially one-dimensional, with only one of the two independent elastic constants of each isotropic phase being used in the model.

The purpose of the present note is to obtain an expression for Young's modulus of a two-component isotropic composite, which takes into account the three-dimensional nature of stress and strain coupling in the material and includes all the elastic constants, to discuss the approximations which have to be used and their significance, and to compare the model with experimental results on some two-component systems.

Model

Consider a system composed of irregularly shaped filler particles (F) embedded in a matrix (M) with different elastic properties, and loaded by a uniaxial, external stress σ in the 3-direction. Neither the six independent components of the stress tensor nor those of the strain tensor can be regarded as spatially constant. However, one can define average stress and strain components for each phase. We assume that the average normal stresses $\bar{\sigma}_i$ and strains $\bar{\epsilon}_i$ $(i = 1,2,3)$ for each phase M and F obey the generalized Hooke's law, as follows:

$$\bar{\epsilon}_1^{M,F} = \frac{1-\nu_{M,F}}{E_{M,F}}\,\bar{\sigma}_1^{M,F} - \frac{\nu_{M,F}}{E_{M,F}}\,\bar{\sigma}_3^{M,F} \tag{3}$$

$$\bar{\epsilon}_3^{M,F} = \frac{-2\nu_{M,F}}{E_{M,F}}\,\bar{\sigma}_1^{M,F} + \frac{1}{E_{M,F}}\,\bar{\sigma}_3^{M,F} \tag{4}$$

$\nu_{M,F}$ are the Poisson's ratios of the two components M and F. The transverse directions 1 and 2 are equivalent to each other. All average shear components are set to zero.

By considering a cross-section of material whose normal is parallel to the 3-direction, it is clear that equation (5) holds:

$$\sigma = \bar{\sigma}_3^F\phi + \bar{\sigma}_3^M(1-\phi) \tag{5}$$

The average longitudinal strains $\bar{\epsilon}_3^{M,F}$ can be related to the composite longitudinal strain ϵ by considering a linear chord through the sample in the 3-direction. The proportion of M or F material encountered is equal to the respective volume fractions. Remembering that the

---

* This work was presented as a paper at The Institute of Materials' Conference POLYMAT'94 held at Imperial College, London in September 1994

† To whom correspondence should be addressed at Polymer and Colloid Group, University of Cambridge, Cavendish Laboratory, Madingley Road, Cambridge, CB3 0HE, UK

strains in the two components are average values, we can thus write:

$$\epsilon = \bar{\epsilon}_3^{\mathrm{F}}\phi + \bar{\epsilon}_3^{\mathrm{M}}(1-\phi) \tag{6}$$

Required ultimately is a relationship between ϵ and σ, where:

$$E = \frac{\sigma}{\epsilon} \tag{7}$$

The next step is to obtain a relationship between the average transverse stresses $\bar{\sigma}_1^{\mathrm{M}}$ and $\bar{\sigma}_1^{\mathrm{F}}$. Since there is no overall external transverse force applied, we may write:

$$\sigma_1 = \bar{\sigma}_1^{\mathrm{F}}\phi + \bar{\sigma}_1^{\mathrm{M}}(1-\phi) = 0 \tag{8}$$

where clearly the sign of the average transverse stresses will be different in each component.

With regard to transverse strains $\bar{\epsilon}_1^{\mathrm{M}}$ and $\bar{\epsilon}_1^{\mathrm{F}}$, we may write:

$$\epsilon_1 = \bar{\epsilon}_1^{\mathrm{F}}\phi + \bar{\epsilon}_1^{\mathrm{M}}(1-\phi) \tag{9}$$

where ϵ_1 is the composite transverse strain.

Let us now examine the series model in the light of the above three-dimensional approach. In order to treat transverse constraints, we consider an arrangement of two components as a unit cube embedded in a homogeneous medium, as shown in *Figure 1*. Assume that the homogeneous medium has the average properties of the composite system, and that the embedded element is constrained to follow this behaviour. In this diagram the two components F and M are coupled in series in the tensile direction 3 and in parallel in the transverse directions 1 and 2. Equations (5) and (9) now take the following form:

$$\bar{\sigma}_3^{\mathrm{M}} = \bar{\sigma}_3^{\mathrm{F}} = \sigma \quad \text{and} \quad \bar{\epsilon}_1^{\mathrm{M}} = \bar{\epsilon}_1^{\mathrm{F}} \tag{10}$$

Combining the above equations we obtain the following expression for the composite Young's modulus E in terms of E_{M}, E_{F}, ν_{M}, ν_{F} and ϕ:

$$\frac{1}{E} = \frac{1-\phi}{E_{\mathrm{M}}} + \frac{\phi}{E_{\mathrm{F}}} - \frac{2\left(\dfrac{\nu_{\mathrm{M}}}{E_{\mathrm{M}}} - \dfrac{\nu_{\mathrm{F}}}{E_{\mathrm{F}}}\right)^2}{\dfrac{1-\nu_{\mathrm{M}}}{(1-\phi)E_{\mathrm{M}}} + \dfrac{1-\nu_{\mathrm{F}}}{\phi E_{\mathrm{F}}}} \tag{11}$$

If both Poisson's ratios are zero ($\nu_{\mathrm{M}} = \nu_{\mathrm{F}} = 0$), equation (11) reduces to the series model or Reuss lower bound (equation (1)), as it must do, since there are no transverse strains. The transverse constraints are thus zero, and the modulus has its lowest possible value.

There is another situation when the Reuss bound is obtained, i.e. when $\nu_{\mathrm{M}}/E_{\mathrm{M}} = \nu_{\mathrm{F}}/E_{\mathrm{F}}$. This arises when the transverse strains in both components are equal without causing any transverse stresses.

For $\nu_{\mathrm{M}} = \nu_{\mathrm{F}} = 0.5$ (both phases incompressible), equation (11) reduces to the rule of mixtures result (equation (2)). All other values of the Poisson's ratios (between 0 and 0.5) lead to modulus values between these two values. *Figure 2* shows some examples.

The parallel model, similarly adapted to this self-consistent three-dimensional scheme, does not lead to anything new, since the rule of mixtures result apertains for all values of Poisson's ratios.

It is clear that this 'longitudinal series/transverse parallel' model as described here does not represent a bound on the Young's modulus, nor is it an exact result, but it may be a useful expression which could describe experimental results satisfactorily. The model will now be compared with experimental results for some particular composites.

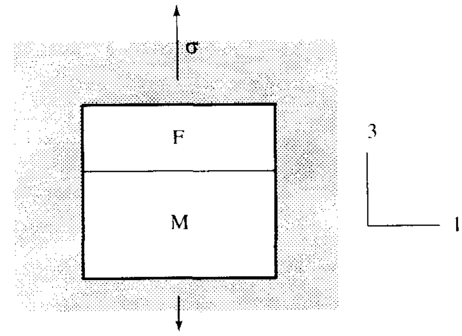

Figure 1 Element of two-component material embedded in average homogeneous material and loaded by a tensile stress σ in the 3-direction (longitudinal series/transverse parallel arrangement)

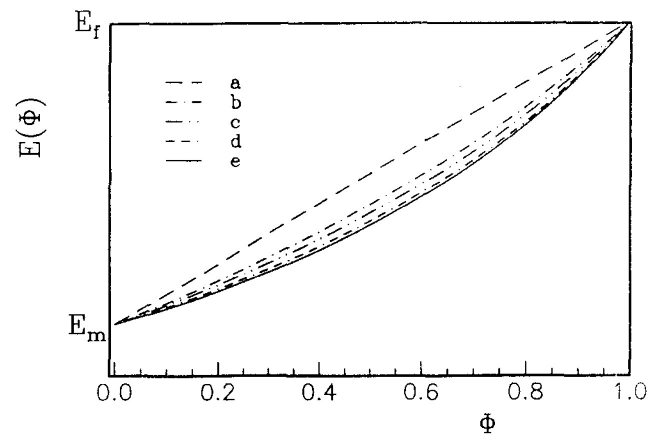

Figure 2 Young's modulus *versus* filler volume fraction ϕ for $\eta = 1$ (equation (11)): (a) $\nu_{\mathrm{M}} = \nu_{\mathrm{F}} = 0.5$; (b)–(d) $0 < \nu_{\mathrm{M}}, \nu_{\mathrm{F}} < 0.5$; (e) $\nu_{\mathrm{M}} = \nu_{\mathrm{F}} = 0$

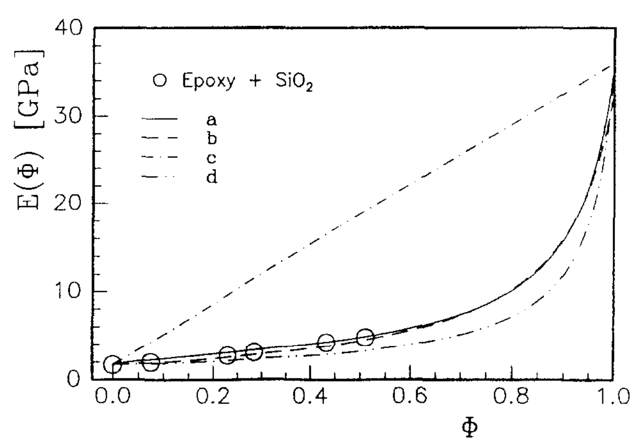

Figure 3 Comparison of Chow's results[1] with: (a) equation (11) of this work; (b) Kerner's model[11]; (c) Reuss bound (equation (1)); (d) rule of mixtures (equation (2))

Comparison of model with experimental results

Figure 3 shows firstly a comparison with the results of Chow[1] on silica-filled epoxy resin. This is a composite system comprising glass spheres embedded in an amorphous resin matrix. The following data for the individual components were used: $E_M = 1.7$ GPa; $\nu_M = 0.35$; $E_F = 36.0$ GPa; $\nu_F = 0.22$. Curve a, which is plotted according to equation (11), gives good agreement with the experimental results (circles). The equation of Kerner's model[11], which was derived for spherical inclusions, also gives reasonable agreement with the data.

Secondly, we applied our model to the behaviour of starch-filled poly-3-hydroxybutyrate (PHB) and a copolymer of PHB and 3-hydroxyvalerate (PHB/HV). These materials have been described elsewhere[16]. This composite system consists of irregularly shaped starch particles of different sizes embedded in a semicrystalline polymer matrix having a spherulitic morphology. The model presented above may be ideally suited to describing the behaviour of such a complex two-component system, because the parameters of the model are simply the average elastic properties of each component, i.e. the two components do not have to be microscopically homogeneous and isotropic.

For this investigation, the Young's modulus of both PHB and PHB/HV (E_M) were known from tensile stress–strain measurements, and the Poisson's ratios (ν_M) had been determined indirectly via the shear modulus G from three-point bending measurements. The Young's modulus of starch (E_F) and its Poisson's ratio (ν_M) were unknown quantities, whose values were required. The values in *Table 1* were used for the matrix elastic constants.

The dashed lines in *Figures 4* and *5* show numerical best fits of equation (11) to the experimental points (circles) where the two parameters E_F and ν_F were allowed to vary freely. A fairly poor fit to the data points is obtained in this case. This discrepancy leads us to examine ways in which the model could be improved. There are no parameters in the model which relate to sample morphology. This implies that, provided the same two components are involved, only the volume fraction determines the resulting modulus. This is not correct in general; Vollenberg and Heikens[2], for example, observed effects due to the size of filler particles. Furthermore, by means of X-ray diffraction measurements Nakamae and co-workers[17] showed that a stress concentration factor of between 2 and 4 occurred in an aluminium particulate epoxy composite, where this factor is defined as the ratio of stress in the filler particles to the applied stress.

There are several more or less unsatisfactory ways of introducing parameters into the above model to account for morphological effects. We choose here to define a stress concentration factor η, given by:

$$\bar{\sigma}_3^F = \eta\sigma \tag{12}$$

This parameter is assumed to be constant, i.e. independent of filler volume fraction ϕ. In addition, the following condition must apply: $\eta\phi < 1$. Furthermore, although the transverse strains $\bar{\epsilon}_1^{F,M}$ are generally not equal, we assume for the present purpose that they are. This leads to the following expression for the composite modulus:

$$\frac{1}{E} = \frac{1-\eta\phi}{E_M} + \frac{\eta\phi}{E_F} + \frac{\left(\frac{-2\nu_M^2}{E_M^2} + \frac{2\nu_M\nu_F}{E_M E_F}\right)\left(\frac{1-\eta\phi}{1-\phi}\right) + \left(\frac{2\nu_M\nu_F}{E_M E_F} - \frac{2\nu_F^2}{E_F^2}\right)\eta}{\frac{1-\nu_M}{(1-\phi)E_M} + \frac{1-\nu_F}{\phi E_F}} \tag{13}$$

Table 1 Values of matrix elastic constants

Matrix M	E (GPa)	G (GPa)	ν
PHB	2.49	0.91	0.37 ± 0.06
PHB/HV	0.61	0.22	0.40 ± 0.05

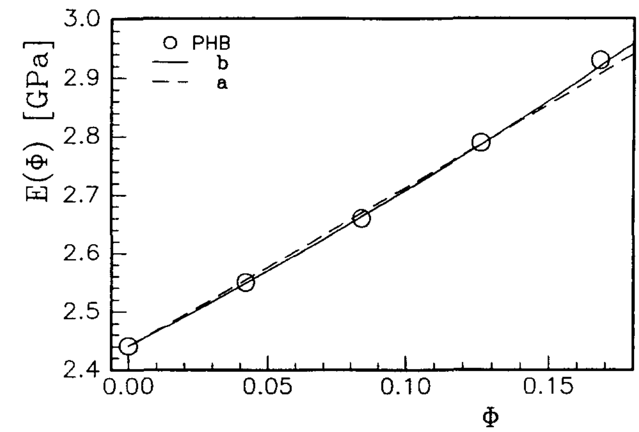

Figure 4 Young's modulus of starch-filled PHB *versus* ϕ: (a) fit according to equation (11) (for $\eta = 1$); (b) fit according to equation (13) ($\eta = 1.4$)

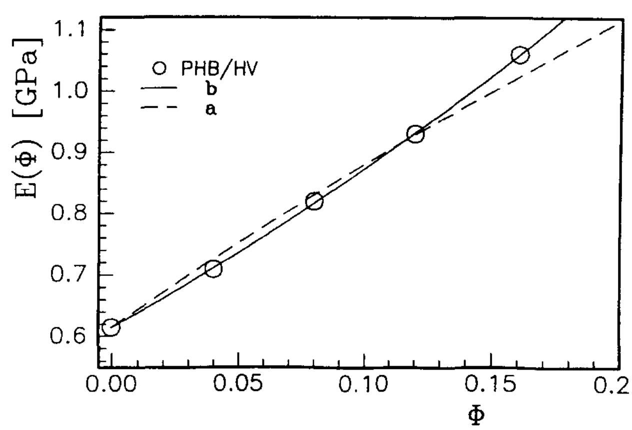

Figure 5 Young's modulus of starch-filled PHB/HV *versus* ϕ: (a) fit according to equation (11) ($\eta = 1$); (b) fit according to equation (13) ($\eta = 1.7$)

Table 2 Young's modulus and Poisson's ratio of starch

	PHB		PHB/HV	
	$\eta = 1$	$\nu = 1.4$	$\eta = 1$	$\eta = 1.7$
E_F (GPa)	6.1	4.9	7.1	5.2
ν_F	0.18	0.19	0.10	0.21

The additional parameter η enables a better fit of the model to the experimental points (see *Figures 4* and *5*), at the same time satisfying a requirement that stress concentrations should be taken into account. *Table 2* shows the values obtained for the Young's modulus of starch and its Poisson's ratio. The stress concentration factor obtained from the best fits is seen to lie between 1 and 2 here, which are plausible values. The estimated Young's modulus of starch is seen to be lower when stress concentrations are considered.

In conclusion, it would be inappropriate to make remarks about the general validity of the proposed model from such limited comparison with experimental results. Further work is still needed in this respect.

References

1 Chow, T. S. *Polymer* 1991, **32**, 29
2 Vollenberg, P. H. T. and Heikens, D. *Polymer* 1989, **30**, 1656
3 Chow, T. S. *J. Polym. Sci., Polym. Phys. Edn.* 1978 **16**, 959
4 Jones, R. M. 'Mechanics of Composite Materials', McGraw-Hill, New York, 1975
5 Arridge, R. G. C. *J. Phys. D: Appl. Phys.* 1975, **8**, 34
6 Boucher, S. *J. Composite Mater.* 1974, **8**, 82
7 Agarwal, B. D. and Broutman, L. J. *Fibre Sci. Technol.* 1974, **7**, 63
8 Lee, K. J., and Westmann, R. A. *J. Composite Mater.* 1970, **4**, 242
9 Hashin, Z. and Shtrikman, S. *J. Mech. Phys. Solids* 1963, **11**, 127
10 Takayanagi, M., Harima, H. and Iwata, Y. *Mem. Fac. Eng. Kyushu Univ.* 1963, **23**, 1
11 Kerner, E. H. *Proc. Phys. Soc. B.* 1956, **69**, 802
12 Arridge, R. G. C. 'An Introduction to Polymer Mechanics', Taylor and Francis, London, 1985
13 Reuss, A. *Z. Angew. Math.* 1929, **9**, 49
14 Voigt, W. 'Lehrbuch der Kristallphysik', Leipzig, 1928
15 Ward, I. M. 'Mechanical Properties of Solid Polymers', 2nd edn, John Wiley, New York, 1983, p. 300
16 Koller, I. and Owen, A. J. *Polym. Int.* in press
17 Nakamae, K., Nishino, T. and Airu, X. *Polymer* 1992, **33**, 2720