Modulus anisotropy of low-density cellular plastics: an aggregate model

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(Received 2 February 1981; revised 22 February 1981)

A structural model of low density (<50 kg m^{-3}) cellular plastics based on an aggregate approach is presented. The model is specifically designed to quantify the modulus anisotropy arising from uniaxial cellular orientation. Relationships are presented which express the five independent foam moduli in terms of the modulus of the solid polymer matrix weighted by the polymer volume fraction and specific orientation parameters. Predictions of the model for isotropic foam systems are discussed and comparisons made with independent experimental data on both the foam Poisson's ratio and density dependence of Young's modulus. A theoretically derived invariant relationship linking the oriented foam Young's moduli to the isotropic value is presented.

INTRODUCTION

Cellular plastic foams are being increasingly used in applications which demand both good insulating and load-bearing properties. A structural understanding of the mechanical properties of such foams is therefore required. Although several structural models have already been proposed[1-7] little attention has been paid to the problem of mechanical anisotropy. Practical foam systems tend to acquire cellular orientation during fabrication which can give rise to large degrees of mechanical anisotropy. A model is presented which attempts to describe the modulus anisotropy of low density cellular plastic foams. The model is restricted to foams with structures which consist of chemically linked solid polymer rod-like struts. A typical example of such a foam is shown in *Figure 1*. Densities below 50 kg m^{-3} are usually required to obtain these structures.

THEORY

The aggregate approach

An aggregate model[8] approach to the problem has been adopted. Such a model views the cellular foam as an aggregate of orienting structural units. For the low-density foams under consideration the structural unit is taken to consist of a solid polymer strut surrounded by a high compliance matrix. To retain the rod-like nature of the struts only foams whose strut lengths are at least three times their width are considered. Foams which possess some intrinsic molecular orientation can be represented by transversely isotropic struts. The matrix in all cases is assumed isotropic. For open cell or reticulated foams the matrix represents the gaseous phase. In closed cell foams the thin polymer windows which radiate from the struts are assumed to decrease the isotropic compliance constants of the matrix. The modified matrix values, however, are still taken to be relatively high with respect to those of the solid polymer strut.

Let an orthogonal frame of reference {1,2,3} be set up within a structural unit such that the 3-axis is parallel to the strut length as shown in *Figure 2*. Let the compliance constants of the polymer strut, matrix and structural unit be S^p_{ij}, S^m_{ij}, S^u_{ij}, respectively.

The S^u_{ij} values are related to S^p_{ij}, S^m_{ij} by making specific mechanical coupling assumptions of continuity of stress or strain. For deformation parallel to the 3-axis a continuity of strain is assumed and for deformation confined solely to the {1,2} plane a continuity of stress is assumed.

Furthermore, by representing the cellular foam structure primarily through the inequality $S^p_{ij} \ll S^m_{ij}$ substantial simplification of the structural unit compliance constants occur:

$$S^u_{33} = \frac{S^p_{33}}{V_f}$$
$$S^u_{22} \simeq S^m_{11}(1 - V_f)$$
$$S^u_{12} \simeq S^m_{12}(1 - V_f)$$

where V_f = volume fraction of polymer strut in a structural unit.

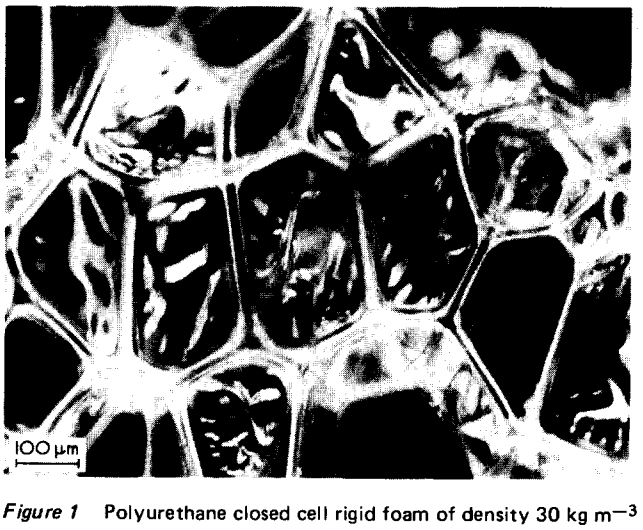

Figure 1 Polyurethane closed cell rigid foam of density 30 kg m^{-3}

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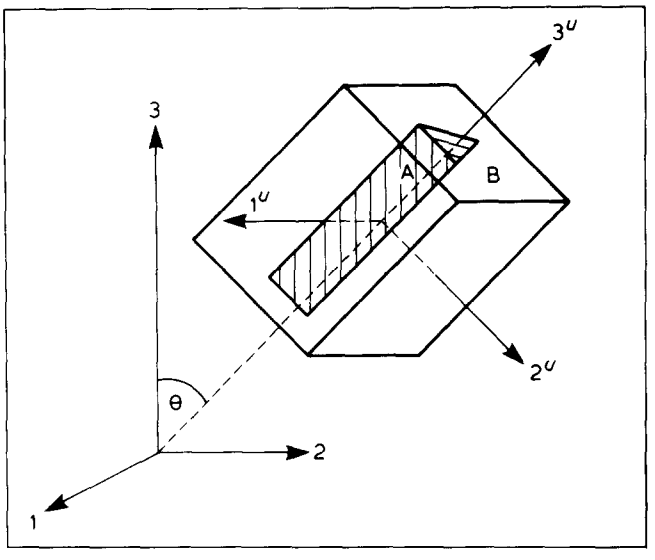

Figure 2 Schematic illustration of a composite structural unit oriented at an angle θ to the principal rise direction of the foam: A, Low compliance solid polymer strut; B, very high compliance sheath representing air for open cell foam or gas plus thin polymer windows for closed cell foam. $\{1,2,3\}$ – Foam co-ordinate frame of reference; $\{1^u,2^u,3^u\}$ – Structural unit coordinate frame of reference

With regard to the shear compliance of S^u_{44}, which involves deformation of the $\{2,3\}$ plane, it is assumed to be dominated by the high compliance matrix, hence:

$$S^u_{44} \simeq S^m_{44}(1-V_f)$$

the compliance constant S^u_{13} can be calculated by considering the Poisson's ratio of the structural unit. If ν^u, ν^p, ν^m, are the Poisson's ratio of the unit, polymer strut and matrix, respectively, then it is assumed that:

$$\nu^u = \nu^p V_f + \nu^m(1-V_f)$$

The gaseous nature of the unit matrix is represented by assuming $\nu^m = 0$, consequently:

$$S^u_{13} = S^p_{13}$$

Determination of the corresponding structural unit stiffnesses C^u_{ij} can be found by matrix inversion, where:

$$S_{ij}C_{jk} = \delta_{ik} \qquad (1)$$

Because of the high compliance values of S^u_{11}, S^u_{22}, S^u_{44}, the matrix inversion leads to the following simple result.

$$C^u_{33} = \frac{1}{S^u_{33}}$$

$$C^u_{11} \simeq C^u_{12} \simeq C^u_{13} \simeq C^u_{14} \simeq 0$$

Table 1 tabulates the final structural unit compliance and stiffness constants assumed for the proposed structural model of low density cellular plastic. The stiffness properties of the structural unit are seen to be represented solely by the Young's modulus of the solid polymer strut.

Let another orthogonal frame of reference $\{1,2,3\}$ be set up within the foam with the 3-axis parallel to the principal orientation direction of the foam as shown in *Figure 2*.

For the assumption of overall foam transverse isotropy the orientation of a structural unit with respect to the foam coordinate frame of reference can be completely defined by the parameter θ, where θ is the angle between the foam 3-axis and the unit 3-axis. The overall foam modulus can now be estimated for any angular distribution of structural units by mechanically coupling such a distribution with a continuity of stress (Reuss average)[9] or a continuity of strain (Voigt average)[10]. For a random aggregate, the Reuss and Voigt values lead to lower and upper bounds respectively[11].

Investigation of solid amorphous polymers indicates that the experimental moduli of both isotropic and oriented samples can be correlated quite well with the average of the Reuss and Voigt bounds[12]. Experimental values close to one or other bound can be obtained by consideration of specific microstructures which weight the mechanical properties towards that bound[13,14]. A common case is that of continuous fibre matt reinforcement (Voigt average) and short fibre reinforcement (Reuss average)[15]. It is proposed that the low density foams under consideration constitute such a weighted structure in which the struts produce a continuous polymer reinforcement of the matrix. A continuity of strain exists from one strut to the next because they can be viewed as a chemically linked continuous fibre matt. It is therefore proposed that the actual foam moduli correspond to the Voigt averaged values of the model.

Let the Voigt averaged foam stiffness and compliance constants be $\langle C_{ij}\rangle^v$ and $\langle S_{ij}\rangle^v$, respectively. Their relationship to the basic structural unit properties can be found by standard 4th rank tensor transformations involving the orientation distribution function of the structural units. A complete list of the full expressions for transversely isotropic systems can be found in ref 8, for example:

$$\langle C_{33}\rangle^v = \langle\sin^4\theta\rangle C^u_{11} + \langle\cos^4\theta\rangle C^u_{33} + 2\langle\sin^2\theta\cos^2\theta\rangle(C^u_{13} + C^u_{44}) \qquad (2)$$

where $\langle f(\theta)\rangle$ is an angular average over all space and over all structural units. Under the particular assumption of the proposed structural model only terms containing C^u_{33} need be considered. Consequently:

$$\begin{aligned}
\langle C_{33}\rangle^v &= \langle\cos^4\theta\rangle C^u_{33}\\
\langle C_{11}\rangle^v &= 3/8\langle\sin^4\theta\rangle C^u_{33}\\
\langle C_{12}\rangle^v &= 1/8\langle\sin^4\theta\rangle C^u_{33}\\
\langle C_{13}\rangle^v &= 1/2\langle\cos^2\theta\sin^2\theta\rangle C^u_{33}\\
\langle C_{44}\rangle^v &= 1/2\langle\cos^2\theta\sin^2\theta\rangle C^u_{33}
\end{aligned}$$

The foam compliance constants $\langle S_{ij}\rangle^v$ can now be determined by matrix inversion (see equation 1). For example:

Table 1 Mechanical constants of the unit of structure

Compliance constants	Stiffness constants
$S^u_{33} = S^p_{33}/V_f$	$C^u_{33} = 1/S^u_{33}$
$S^u_{11} \to \infty$	$C^u_{11} = 0$
$S^u_{13} = S^p_{13}$	$C^u_{13} = 0$
$S^u_{12} \to \infty$	$C^u_{12} = 0$
$S^u_{44} \to \infty$	$C^u_{44} = 0$

$$\langle S_{33}\rangle^v = \frac{1}{C^u{}_{33}\left[\langle\cos^4\theta\rangle - \dfrac{\langle\sin^2\theta\cos^2\theta\rangle}{\langle\sin^4\theta\rangle}\right]} \quad (3)$$

ISOTROPIC CASE

For the isotropic case the angular averages can be precisely evaluated by random integration over all space giving:

$$\langle\sin^4\theta\rangle = 8/15;\ \langle\cos^4\theta\rangle = 3/15;\ \langle\sin^2\theta\cos^2\theta\rangle = 2/15$$

Equation (3) therefore reduces to:

$$\langle S_{33}\rangle^v = \frac{6}{C_{33}{}^u} = \frac{6S_{33}{}^p}{V_f}$$

The predicted foam modulii $M^f{}_{ij}$ are just the reciprocals of the Voigt averaged compliances:

$$M^f{}_{ij} = \frac{1}{\langle S_{ij}\rangle^v}$$

The Young's modulus of the foam $M^f{}_{33}$ is therefore:

$$M^f{}_{33} = 1/6\, M^p{}_{33}\, V_f \quad (4)$$

Similarly it can be shown that:

$$M^f{}_{44} = 1/15\, M^p{}_{33}\, V_f \quad (5)$$

The foam modulus is therefore reduced not only by the simple volume fraction dependence but also by the 1/6 coefficient. The 1/6 dependence arises from the dependence of the modulus on 4th order orientation parameters. Thus, only 1/6 of the struts are oriented close enough to the measurement direction to be effective load bearers.

Poisson's ratio

Prediction of the Poisson's ratio tends to be a sensitive test for any model since deformations in two mutually perpendicular directions are required. Since v is defined in the isotropic case as:

$$v = 1/2\left(\frac{M_{33}}{M_{44}}\right) - 1$$

Substitution of equations (4) and (5) give:

$$v^{\text{foam}} = 1/4$$

ANISOTROPY RELATIONSHIPS

Most practical foam systems have an upper anisotropy limit of $M_{33}/M_{11} \simeq 3$. It is assumed that within this orientation range the identification of the Voigt average as the actual foam modulus is still valid. Anisotropy equations can therefore be defined as shown by equation (3).

Introduction of a specific deformation model at this stage, such as an affine deformation for example, would allow a precise evaluation of the relevant deformation functions to be made. Irrespective, however, of the precise nature of the deformation involved, the necessary decrease of the averaged angle θ for a transversely isotropic deformation leads to reasonable assumptions regarding the relative change of magnitude of the orientation functions during the deformation process. From equation (3), for instance, it is assumed that:

$$\langle\cos^4\theta\rangle \gg \frac{\langle\sin^2\theta\cos^2\theta\rangle^2}{\langle\sin^4\theta\rangle}$$

giving a reduction to the approximate relationship:

$$M^f{}_{33} = \langle\cos^4\theta\rangle\, M^p{}_{33}\, V_f \quad (6)$$

Similar approximation procedures give:

$$M^f{}_{11} = 1/3\langle\sin^4\theta\rangle\, M^p{}_{33}\, V_f \quad (7)$$

$$M^f{}_{12} = -\langle sin^4\theta\rangle\, M^p{}_{33}\, V_f \quad (8)$$

$$M^f{}_{13} = -\frac{\langle\sin^4\theta\rangle\langle\cos^4\theta\rangle}{\langle\sin^2\theta\cos^2\theta\rangle}\, M^p{}_{33}\, V_f \quad (9)$$

$$M^f{}_{44} = 1/2\langle\sin^2\theta\cos^2\theta\rangle\, M^p{}_{33}\, V_f \quad (10)$$

For a foam of some arbitary level of orientation therefore, equations (6) and (7) can be combined to form an invariant relationship equating the oriented modulus anisotropy to the isotropic modulus provided the following approximation is assumed to hold over the practical orientation range considered.

$$\langle\cos^4\theta\rangle + \langle\sin^4\theta\rangle = K$$

This allows equations (6) and (7) to give:

$$M^f{}_{33} + 3M^f{}_{11} = K\, M^p{}_{33}\, V_f$$

Using equation (4):

$$M^f{}_{33} + 3M^f{}_{11} = 6K\, M^f{}_{\text{isotropic}} \quad (11)$$

(For an affine deformation $K \simeq 3/4$).

DISCUSSION

A comprehensive model of the modulus anisotropy of low-density cellular plastic foams has been proposed. The model is restricted to cellular structures in which most of the solid polymer resides in chemically-linked struts. Foams with such structures tend to have densities less than 50 kg m^{-3}. For open cell or reticulated foams the volume fraction of polymer can be simply replaced by the density ratio ρ^f/ρ^p where ρ^f and ρ^p are the density of the foam and solid polymer, respectively.

For closed cell foams the polymer residing in the windows whilst not, according to the model, influencing the foam modulus in a load-bearing sense, cannot be neglected in any calculation of V_f. Consequently simple replacement of V_f by the foam density ratio should lead to qualitative agreement only.

The principal predictive claims of the theoretical model are contained in equations (6)–(11). These define the entire modulus anisotropy of oriented low density foams. The deformation is solely in terms of the Young's modulus of

the solid polymer struts, their volume fraction composition and specific orientation parameters. These parameters take the form of angular averages of the orientation of the struts with respect to the direction of foam modulus measurement.

Although the mechanical anisotropy of low density cellular plastics is of major importance in the production of products of acceptable performance, surprisingly little published work exists on the experimental characterization of such anisotropy. Several independent experimental studies[2,4,17] have, however, been performed on essentially isotropic, open-cell materials and in this area it is possible to check the predictions of the model. All the studies show that the Young's modulus of the foam at low densities can be related to the intrinsic Young's modulus of the solid polymer by a relationship identical to equation (4). Similarly, the comprehensive experimental investigation of cellular plastic Poisson's ratios by Rinde[16] also gives excellent agreement with the theoretically predicted value of 1/4.

The proposed model attempts to describe only the initial portion of the modulus/density relationship. For example, the Young's modulus/density dependence for cellular plastics, when examined over an extreme range of densities, takes on a curved form[2,7] of increasing gradient with respect to density.

This curved relationship is viewed in the light of the model as a consequence of distinct shifts in the structural nature of the foam. Initially, the aggregate of rod-like struts allows only a 1/6 gradient resulting from their restricted uniaxial contributions. At higher densities the foam structure becomes more representative of an aggregate of plates. Since the plates confer a planar contribution on the composite modulus an enhanced gradient dependence is obtained. At densities near to the value for the solid polymer matrix the gradient becomes unity (law of mixtures). This shift to increasing levels of structural efficiency results in the empirically determined curve.

ACKNOWLEDGEMENTS

The author wishes to thank M. Koval for the writing of all necessary computer programs during the development of the model.

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