1. Introduction

Van der Poel’s method [1] for calculating the shear modulus of a particulate composite is an excellent method capable of giving accurate results [1, 2], but because of several disadvantages has not been widely used. In the original presentation the calculation was complicated. A table of values was therefore provided, but this table was limited to materials for which Poisson’s ratio of the matrix was 0.5. In addition, there was an error in the derivation [3], and its effect on the results was not known. Recently, however, van der Poel’s method has been reexamined [4], the error corrected, and the method extended for use with matrix materials having any value for Poisson’s ratio. The effect of the error was shown to be minor, and a new table of values was provided. Continued studies have now resulted in an improved understanding of the theory, and have provided significant simplifications in the calculation. This work is presented here.

2. Review of the Method

A detailed derivation based upon a method developed by Frölich and Sack [5] has been given elsewhere [4]. Consider an idealized composite material consisting of small spheres imbedded in a matrix. The spheres are of approximately the same size, are firmly attached to the matrix, and are uniformly distributed so that the composite material is macroscopically homogeneous and isotropic. To simplify calculations consider a unit length chosen such that on the average there is one sphere of filler in each spherical volume $4\pi/3$ of unit radius. If $a$ is the radius of the filler sphere, then by definition of the unit radius, $a^3$ is equal to the volume fraction $\phi$ of the filler.

Consider a large sphere of homogeneous material. Consider also a second sphere of the same size consisting mostly of the homogeneous material but having the following structure in the central region: A filler sphere of radius $a$ is located at the origin. Surrounding this out to a radius unity is a shell of matrix material, which in turn is imbedded in the homogeneous material. The mechanical properties of this homogeneous material are assumed to be the same as the average macroscopic properties that are sought for the composite material considered here.

If the same boundary stresses are applied to each of the spheres, it is assumed that the displacements in the two spheres will be the same at a distance $r \gg 1$, except for terms of a high order in $1/r$. Thus it is required that

Displacement at $r \gg 1$ in second sphere =
\[
\text{Displacement at } r \gg 1 \text{ in first sphere } = 1 + \sum_{n=3} \frac{\text{Const}.}{r^n} \quad (1)
\]

Let the first sphere be subjected at the boundary to the deviatoric stress system

\[
\sigma_r = 2TP_2(\cos \theta), \quad \tau_{r\theta} = TP_2(\cos \theta), \quad \tau_{r\phi} = 0. \quad (2)
\]
where $\sigma_r$ is the radial tensile stress, and $\tau_{r\theta}$ and $\tau_{r\phi}$ are the tangential shear stresses on the spherical boundary surface at radius $r$. The corresponding boundary displacements in the $r$, $\theta$, and $\phi$ directions are

$$u_r = \frac{Tr}{G} P_2(\cos \theta), \quad u_\theta = \frac{Tr}{2G} P'_2(\cos \theta), \quad u_\phi = 0. \quad (3)$$

In the above equations $T$ is a stress magnitude, and $G$ is the shear or rigidity modulus. $P_2(\cos \theta)$ is the second Legendre Polynomial, and $P'_2(\cos \theta)$ is its derivative with respect to $\theta$.

The stresses and displacements at any radius $r$ within sphere 1 of the homogeneous material are also given by eqs (2, 3), but the stresses and displacements within sphere 2 having the special structure must be found by solving Laplace’s equations. These solutions subject to the requirement of symmetry about the $\theta = 0$, axis, and the requirement that the solutions possess the same $P_2$ symmetry as those of eqs (2, 3), have been derived in the previous paper [4] and are given by

$$u_r = \left[ A_mr + B_mr^{-4} + C_mr^{-3} + D_mr^{-2} \right] P_2(\cos \theta) \quad (4)$$

$$u_\theta = \left[ \frac{1}{2} A_mr - \frac{1}{3} B_mr^{-4} + \frac{7}{12} \frac{C_mr^{-3}}{\nu_m} + \frac{1}{5} \frac{D_mr^{-2}}{\nu_m} \right] P'_2(\cos \theta) \quad (5)$$

$$\sigma_r = 2G_m \left[ A_m - 4B_mr^{-5} - \frac{1}{2} \frac{C_mr^{-2}}{\nu_m} \right] P_2(\cos \theta) - \frac{2(5 - \nu_m)}{5 - 4\nu_m} D_mr^{-3} \quad (6)$$

$$\tau_{r\theta} = G_m \left[ A_m + \frac{8}{3} B_mr^{-5} + \frac{7 + 2\nu_m}{6\nu_m} C_mr^{-2} + \frac{2(1 + \nu_m)}{5 - 4\nu_m} D_mr^{-3} \right] P'_2(\cos \theta) \quad (7)$$

where the quantity $\nu$ is Poisson’s ratio.

In the above solutions the subscript $m$ refers to the matrix medium of the region $a \leq r \leq 1$ in which these equations hold. Similar equations holding within the filler sphere $r \leq a$ are designated by the subscript $f$, but in order that these solutions be finite at the origin the coefficients $B_f$ and $D_f$ are set equal to zero. For solutions holding in the region $r \geq 1$ outside the matrix shell no subscripts are used. In these solutions the coefficients $C$ and $D$ do not appear because of the requirement eq (1), and the coefficient $A$ is set equal to $T/G$ for consistency with the applied boundary stresses, eq (2).

At the boundaries $r=a$ and $r=1$, continuity of stress and displacement is required. This results in a system of eight equations in the seven unknowns $A_f$, $C_f$, $A_m$, $B_m$, $C_m$, $D_m$, $B$, assuming for the moment that the shear modulus $G$ is known. If $T$ is considered as an additional unknown, a system of linear homogeneous equations results. In order that the solution of this system be nontrivial, or that $A_f$, $C_f$, $A_m$, etc. can be expressed in terms of the quantity $T$, the determinant of the matrix of coefficients must vanish. Setting this determinant equal to zero evaluates $G$, the shear modulus of the homogeneous material and the quantity sought, in terms of the known quantities $G_f$, $G_m$, $\nu_f$, $\nu_m$.

### 3. Results

The determinantal equation for $G$ can be reduced to the form

$$\alpha X^2 + \beta X + \gamma = 0. \quad (8)$$

where

$$\alpha = \left[ 4P(7 - 10\nu_m) - Sa^2 \right] \left[ Q - (8 - 10\nu_m) (M - 1) a^2 \right] - 126P(M - 1)a^2 (1 - a^2)^2 \quad (9)$$

$$\beta = 35(1 - \nu_m)P[Q - (8 - 10\nu_m) (M - 1) a^2] - 15(1 - \nu_m) [4P(7 - 10\nu_m) - Sa^2] (M - 1) a^3 \quad (10)$$

$$\gamma = -525P(1 - \nu_m)^2 (M - 1) a^3 \quad (11)$$

and the quantities $P$, $Q$, and $S$ are defined as

$$P = (7 + 5\nu_f)M + 4(7 - 10\nu_f) \quad (12)$$

$$Q = (8 - 10\nu_m)M + 7 - 5\nu_m \quad (13)$$

$$S = 35(7 + 5\nu_f)M (1 - \nu_m) - P(7 + 5\nu_m) \quad (14)$$

$M$ is the ratio $G_f/G_m$, and the quantity $X$ is equal to $(G/G_m)^{-1}$. Equation (8) has two real roots, one of which is negative and extraneous. The positive root provides the required value for $G$.

Equations (8) through (11), although more compact than those given previously [4] are still unwieldy, so it becomes worthwhile to search for an approximate solution. Such a solution could be obtained as a series expansion in powers of $a = \varphi^{1/3}$, but in order to obtain sufficient accuracy too many terms would be required. A more compact solution is one of the Padé type:

$$X = \frac{n_0 + n_1 \lambda + n_2 \lambda^2 + \cdots}{1 + d_1 \lambda + d_2 \lambda^2 + \cdots} \quad (15)$$

where the parameter $\lambda$ is given by

$$\lambda = \frac{(M - 1)a^3}{Q} = \frac{(M - 1)\varphi}{Q} \quad (16)$$

To obtain this approximate solution $\lambda Q$ is substituted for $(M - 1)a^3$ in eqs (9), (10), (11) for $\alpha$, $\beta$ and $\gamma$, and these equations are then substituted together with eq (15) into eq (8). The unknown constants $n_1$, $n_2$, $d_1$, $d_2$, etc. are evaluated by equating to zero the coeffi-
cients of \( \lambda \) in the resulting expression. Several different expansions of the form eq (15) can be obtained, because the pairs of constants \( n_2, d_1; n_1, d_2; \) etc. are interrelated. Thus one of the pair members is arbitrary and may be set equal to zero. A suitable approximate expression is

\[
X_4 = \frac{15(1 - \nu_m)\lambda + 810(1 - \nu_m)(1 - \alpha^2)^2\lambda^3}{1 - (8 - 10\nu_m)\lambda - 108(1 - \alpha^2)(8 - 10\nu_m - 3R/14P)\lambda^3}
\]

(17)

where \( R \) is given by

\[
R = 4P(7 - 10\nu_m) - Sa'.
\]

(18)

To a first approximation eq (17) becomes

\[
X_1 = \frac{15(1 - \nu_m)(M - 1)\varphi}{(8 - 10\nu_m)M + 7 - 5\nu_m}.
\]

(19)

This approximation has been given previously by Hashin [6], and by Eshelby [7], who found it by different methods. It may be regarded as a generalized form of Smallwood’s equation [8].

To a second approximation eq (17) becomes

\[
X_2 = \frac{15(1 - \nu_m)(M - 1)\varphi}{Q - (8 - 10\nu_m)(M - 1)\varphi}.
\]

(20)

It can be shown by algebraic manipulation that this approximation is the same as Kernér’s equation [9], or Hashin and Shtrikman’s equation for the highest lower bound [10].

Equation (20) provides a means of further simplifying the exact calculation of \( X \). If the quantities \( \alpha, \beta, \gamma \) are multiplied by the factor

\[
15(1 - \nu_m)/[Q - (8 - 10\nu_m)(M - 1)\alpha^3],
\]

the following new quantities \( \alpha', \beta', \gamma' \) are obtained

\[
\alpha' = 15(1 - \nu_m)R - 126P(1 - \alpha^2)^2X_2
\]

(21)

\[
\beta' = 525P(1 - \nu_m)^2 - 15(1 - \nu_m)RX_2
\]

(22)

\[
\gamma' = -525P(1 - \nu_m)^2X_2
\]

(23)

These new quantities \( \alpha', \beta', \gamma' \) may be substituted into eq (8) to calculate \( X \).

The approximate solution eq (20) can be used with Newton’s method to obtain a better approximation. Let

\[
F(X) = \alpha'X^2 + \beta'X + \gamma'
\]

\[
= \alpha' \left( X + \frac{\beta'}{2\alpha'} \right)^2 - \left( \frac{\beta'}{4\alpha'} - \gamma' \right)
\]

(24)

Then to a first approximation

\[
F'(X) = F(X_2) + (X_2 - X)F'(X_2) = 0.
\]

(25)

or

\[
X = X_2 - \frac{F(X_2)}{F'(X_2)}
\]

(26)

Evaluating the quantities \( F(X_2) \) and \( F'(X_2) \) gives

\[
X = \frac{126P(1 - \alpha^2)^2X_2^3}{525P(1 - \nu_m)^2 + 15(1 - \nu_m)RX_2 - 252P(1 - \alpha^2)^2X_2^3}
\]

(27)

The function \( F(X) \) is a parabola symmetric about the axis \( X = -\beta'/2\alpha' \). At the point where \( F(X) \) intersects this axis \( F(X) \) is negative. As \( X \) increases positively \( F(X) \) becomes less negative and is zero where it intersects the \( X \) axis. The value of \( F(X_2) \) is

\[
F(X_2) = -126P(1 - \alpha^2)^2X_2^3
\]

(28)

and is negative, so that eq (20) provides a value of \( G \) less than the true value. On the other hand the value of \( F(X_\infty) \) is

\[
F(X_\infty) = \alpha'(X_\infty - X_2)^2
\]

(29)

and is positive. If \( X_\infty \) is a sufficiently good approximation, \( X_\infty \) will be an improved approximation, slightly exceeding the true value.

4. Discussion

The exact solution of eq (8) and the approximate solutions eqs (17) and (27) have been studied for volume fractions of filler \( \varphi \) between 0 and 1.0, using the following values for the parameters \( \nu_f, \nu_m, \) and \( M \):

\[
\nu_f = 0.1; \quad \nu_m = 0.4; \quad M = 30, 100
\]

\[
\nu_f = 0.1; \quad \nu_m = 0.5; \quad M = 1000
\]

\[
\nu_f = 0.25; \quad \nu_m = 0.3; \quad M = 10
\]

\[
\nu_f = 0.25; \quad \nu_m = 0.4; \quad M = 30, 100
\]

\[
\nu_f = 0.25; \quad \nu_m = 0.5; \quad M = 100, 10^3, 10^4, 7 \times 10^4, \infty
\]

\[
\nu_f = 0.35; \quad \nu_m = 0.4; \quad M = 30, 100
\]

\[
\nu_f = 0.35; \quad \nu_m = 0.5; \quad M = 1000.
\]

![Plot of relative shear modulus G/G_m versus volume fraction of filler for the case G_dG_m = 30, \nu_f = 0.25, \nu_m = 0.40.](image)
Plots selected from these calculations are given for two representative composite materials in figures 1 and 2. In figure 1 the dependence of relative shear modulus $G/G_m$ on the volume fraction of filler $\varphi$ is shown, as calculated from eqs $(8), (17)$ and $(27)$. The parameter values used were $\nu_f = 0.25, \nu_m = 0.4$ and $M = 30$, and are representative of a composite consisting of small glass spheres imbedded in a rigid epoxy matrix. For this example it is seen that the plots for the $X_4$ and $X_1$ approximations lie reasonably close to either side of the plot for the exact value, but that the plotted values for the $X_2$ approximation are too low when the volume fraction $\varphi$ exceeds 0.4.

Figure 2 is a similar plot of values calculated using the parameter values $\nu_f = 0.25, \nu_m = 0.50$ and $M = 70,000$. These values are representative of a composite consisting of small glass spheres imbedded in a matrix of lightly vulcanized rubber. In this example the $X_2$ approximation calculated from eq $(27)$ gives values close to the exact values calculated from eq $(8)$, when $\varphi$ is less than 0.4. The approximate values $X_2, X_3, X_4$ are good only for values of $\varphi$ up to 0.2.

Equation $(16)$ for the parameter $\lambda$ may be expressed in the following form

\[
\lambda = \frac{\varphi}{8 - 10\nu_m + 15(1 - \nu_m)} \frac{M}{(M - 1)}
\]  

When $\nu_m = 0.4, M = 30, \varphi = 0.4$, the value of $\lambda$ is $\lambda = 0.09$, and when $\nu_m = 0.5, M = 70,000, \varphi = 0.4$, the value of $\lambda$ is $\lambda = 0.13$. Thus for values of $\varphi$ greater than 0.4 the terms in eq $(17)$ representing the successive approximations $X_2, X_3, X_4$ are relatively large, indicating that this series solution does not converge very rapidly, as evidenced in figures 1 and 2. The approximations provided by eq $(17)$ are probably reliable only for values of $\varphi$ up to 0.2. In this connection it is appropriate to recall that the approximation $X_2$ is equivalent to the widely used equation of Kerner [9], or Hashin and Shtrikman’s equation for the highest lower bound [10].

It has been pointed out previously that values of $G/G_m$ calculated from van der Poel’s equation are insensitive to values of $\nu_f$. The extent of this insensitivity can be seen from eq $(17)$. The values of $R$ and $P$ are moderately dependent on the values of $\nu_f$, and these quantities first appear in the coefficient of $\lambda^3$ in the denominator of eq $(17)$. In other words the approximation $X_4$ is the first one to be affected by the value of $\nu_f$.

Although in the examples of figures 1 and 2 the approximation of eq $(27)$ gives reasonably accurate values, there are situations when eq $(27)$ fails because the approximation $X_2$ is not good enough. This occurs when for a sufficiently high value of $\varphi$ the coefficient $\beta'$, eq $(22)$, changes from a positive to a negative value. The axis of the parabola $F(X) = 0$, then intersects the $X$ axis at the positive point $X = (-\beta')/2\alpha'$. This point will usually be near $X_2$, and as $\varphi$ increases will exceed it. In either event the slope $F'(X_2)$ will be too low or even negative, and the calculated value of $X_3$ will be grossly inaccurate.

In the studies using the parameter values listed above, it was found that $\alpha'$ was always positive and $\gamma'$ was always negative, but when $\varphi$ exceeded 0.5, $\beta'$ changed sign and became negative. For values of $\varphi$ exceeding 0.5 the values of $X_3$ were reasonably accurate when $M$ was less than 100, but when $M$ exceeded 100 the values of $X_3$ were grossly inaccurate or negative.

It is fortunate that eq $(27)$ gives a good approximation for values of $\varphi$ up to 0.4 or 0.5. At values of $\varphi$ greater than 0.5 the assumptions upon which the theory is based are no longer valid. For instance, the packing fraction of spheres of equal size arranged on a cubic lattice is 0.52, and the packing fraction of equal size spheres in hexagonal close packed arrangement is 0.74. As a practical matter it is difficult to prepare a particulate composite material with a volume fraction of filler $\varphi$ exceeding 0.5 without introducing excessive void content. However Schwarzl [2] and van der Poel [1] report that the theory agrees well with experimental data for values of $\varphi$ up to 0.5. It seems then that the calculations of the theory have an empirical validity beyond the theoretical expectations.
5. References

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