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COMPLEX MODULI OF VISCOELASTIC COMPOSITES—I. GENERAL THEORY AND APPLICATION TO PARTICULATE COMPOSITES†

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Abstract—A correspondence principle is developed by means of which effective complex moduli of viscoelastic composites can be determined on the basis of analytical expressions for effective elastic moduli of composites. The method is applied to determine complex moduli of isotropic particulate composites with special attention to the composite spheres assemblage model. Very good agreement with published experimental results for effective complex moduli is obtained.

1. INTRODUCTION

A RATIONAL theory of the quasi-static viscoelastic behavior of statistically isotropic composites has first been given in [1] and of fiber reinforced materials in [2]. The basic tool in the analysis was the development of a correspondence principle which relates effective elastic moduli of composites to effective relaxation moduli and creep compliances of viscoelastic composites.

The purpose of the present work is to investigate the macro-dynamic behavior of viscoelastic composites in terms of effective complex moduli. As is well known, analysis of dynamic behavior of homogeneous viscoelastic materials is greatly facilitated by the introduction of the concept of complex viscoelastic moduli. It will be seen that within certain restrictions it is also possible to define effective complex moduli of viscoelastic composites. These effective complex moduli are also found to be related to the effective elastic moduli of composites by a correspondence principle which is simpler to use than the one mentioned above.

2. GENERAL THEORY

It has been shown in [1] that effective relaxation moduli of statistically homogeneous viscoelastic composites may be defined by

$$\bar{\sigma}_{ij}(t) = \int_{-\infty}^{t} C^*_{ijkl}(t-\tau)\frac{\partial \bar{\varepsilon}_{kl}(\tau)}{\partial \tau}\mathrm{d}\tau \tag{2.1}$$

where σ_{ij} and ε_{ij} are the stress and strain tensors, respectively, an overbar denotes local or global average of a statistically homogeneous field, C^*_{ijkl} is the effective relaxation moduli tensor and t is time. Here and in the following the range of subscripts is 1, 2, 3 and repeated subscripts denote summation over their ranges.

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Suppose that the composite is subjected to an average sinusoidal strain

$$\bar{\varepsilon}_{ij}(t) = \tilde{\varepsilon}_{ij}\, e^{i\omega t}, \tag{2.2}$$

where ω is the circular frequency, $i = \sqrt{(-1)}$ and $\tilde{\varepsilon}_{ij}$ are time independent but may be complex and functions of ω. Introduction of (2.2) into (2.1) yields

$$\bar{\sigma}_{ij}(t) = \tilde{\varepsilon}^0_{kl} i\omega \int_{-\infty}^{t} C^*_{ijkl}(t-\tau) e^{i\omega\tau}\, d\tau. \tag{2.3}$$

The change of variable

$$t - \tau = u,$$

in (2.3) yields

$$\bar{\sigma}_{ij}(t) = \tilde{\varepsilon}^0_{kl} i\omega\, e^{i\omega t} \int_0^{\infty} C^*_{ijkl}(u)\, e^{-i\omega u}\, du. \tag{2.4}$$

Let

$$B^*_{ijkl}(i\omega) = i\omega \int_0^{\infty} C^*_{ijkl}(u)\, e^{-i\omega u}\, du. \tag{2.5}$$

Then (2.4) can be rewritten in the form

$$\bar{\sigma}_{ij}(t) = \tilde{\sigma}_{ij}\, e^{i\omega t} \tag{2.6}$$

$$\tilde{\sigma}_{ij} = B^*_{ijkl}(i\omega)\tilde{\varepsilon}_{kl}. \tag{2.7}$$

Equations (2.6–7) show that $\bar{\sigma}_{ij}(t)$ is now also in periodic sinusoidal form.

Because of the formal resemblance of (2.7) to an elastic stress strain law the tensor components B^*_{ijkl} are called *effective complex moduli.* It is seen that they relate uniquely the amplitudes of sinusoidal stress and strain vibrations. Since the quantities in (2.7) are in general complex the stress and strain vibrations for same frequency ω are not in phase.

Evidently, B^*_{ijkl} are i, j and k, l symmetric. They are also ij, kl symmetric if it is assumed that the local $C_{ijkl}(t)$ are ij, kl symmetric.

Whenever convenient B^*_{ijkl} will be separated into real and imaginary parts. Thus

$$B^*_{ijkl}(i\omega) = B^{*R}_{ijkl}(\omega) + i B^{*I}_{ijkl}(\omega). \tag{2.8}$$

The present derivation of effective complex moduli of composites is in every sense analogous to similar derivations for complex moduli of homogeneous viscoelastic materials.

It should be noted that the effective complex moduli as defined by (2.5) involve a one sided Fourier transform which need not exist. This formal difficulty is not peculiar to composites, it also arises in similar definitions of complex moduli of homogeneous viscoelastic materials and may be resolved in similar fashion in both cases, see e.g. Gross [3].

At this point it is necessary to consider a difficulty which arises because of material heterogeneity. It should be recalled that the effective stress strain relation (2.1) is valid in the strict sense only for spatially statistically homogeneous fields of stress and strain. A device for producing such stress and strain fields in heterogeneous bodies is the imposition of so-called homogeneous boundary conditions on the surface of a large composite. These are boundary conditions which in *homogeneous* bodies produce *homogeneous* states of stress and strain, see [1]. However, dynamic equations of homogeneous elastic or viscoelastic bodies can not be satisfied by spatially uniform fields of stress and strain because of the appearance of inertia terms. It can therefore not be expected that in vibrations of heterogeneous, albeit statistically homogeneous, bodies there can exist spatially

statistically homogeneous states of stress and strain, for homogeneity is a special case of statistical homogeneity. It seems that in order to salvage the development leading from (2.1) to (2.11) it must be assumed that the averages in (2.1) are local averages over representative volume elements (RVE). As the position of the RVE varies in the space of the body so does the local average. However, the gradients may be assumed to be sufficiently small to permit the assumption that the stress and strain fields are *locally statistically homogeneous*, thus permitting retention of (2.1) locally. It is intuitively clear that such assumptions will be quite accurate for low vibration frequencies (large wave-lengths) whereas for high frequencies they may lead to serious errors.

Suppose that $\bar{u}_i(\mathbf{x}, t)$ is the local RVE displacement average, where $\mathbf{x}$ specifies the position of the RVE (e.g. its centroid). The average $\bar{u}_i(\mathbf{x}, t)$ may be called the macro-displacement. We may similarly define a macro-stress $\bar{\sigma}_{ij}(\mathbf{x}, t)$. The macro-strain $\bar{\varepsilon}_{ij}(\mathbf{x}, t)$ is defined by

$$\bar{\varepsilon}_{ij} = \tfrac{1}{2}(\bar{u}_{i,j} + \bar{u}_{j,i}). \tag{2.9}$$

The assumption of local statistical homogeneity implies that for an elastic composite

$$\bar{\sigma}_{ij}(\mathbf{x}, t) = {}^{e}C^{*}_{ijkl}\bar{\varepsilon}_{ij}(\mathbf{x}, t) \tag{2.10}$$

where ${}^{e}C^{*}_{ijkl}$ are the usual effective elastic moduli. For a viscoelastic composite (2.10) is replaced by (2.1), which then assumes the form (2.6–7) for sinusoidal time variation. It is seen that $\bar{\sigma}_{ij}$ and $\bar{\varepsilon}_{ij}$ in (2.2–7) may be interpreted as the macro-stresses and macro-strains which have been defined above.

A theory based on the assumptions (2.10) and (2.1), (2.6–7) for relation between macro-stresses and macro-strains may be called: *first approximation* to macro-mechanics of composites. It may be shown that for elastic or viscoelastic *two phase* materials the first approximation leads to a mathematical formulation in terms of macro-variables which is mathematically similar to the usual classical formulations for homogeneous bodies. In the first order macro-theory the effective elastic moduli replace the usual homogeneous elastic moduli and it is necessary to replace the usual density by an *effective density* which is not in general equal to the average density of the composite. Details will be published elsewhere.

The problem of dynamics of composites without the assumption of local statistical homogeneity is a most important one. Investigation has begun only very recently, e.g. [4, 5], for elastic composites. Here we shall only be concerned with the first approximation as described above. It should however be borne in mind that given the polycrystalline and otherwise heterogeneous nature of materials to which classical dynamic continuum theory is applied with good experimental verification, it would seem that such an approximation should be valid for a respectable range of frequency.

Having provided some physical justification for the concept of effective complex moduli we proceed to derive a correspondence principle which will enable us to determine effective complex moduli on the basis of effective elastic moduli expressions.

Consider the Laplace Transform (LT) of the effective relaxation moduli

$$\hat{C}^{*}_{ijkl}(p) = \int_0^\infty C^{*}_{ijkl}(t)\, \mathrm{e}^{-pt}\, \mathrm{d}t. \tag{2.11}$$

Then the LT of (2.1) is given by the convolution theorem in the form

$$\hat{\bar{\sigma}}_{ij}(p) = \Gamma^*_{ijkl}(p)\hat{\bar{\varepsilon}}_{kl}(p) \tag{2.12}$$

where

$$\Gamma^*_{ijkl}(p) = p\hat{C}^*_{ijkl}(p). \tag{2.13}$$

The tensor components Γ^*_{ijkl} have been given the name transform domain (TD) effective moduli in [1]. The correspondence principle for elastic and viscoelastic heterogeneous media as developed in [1] gives a relation between effective TD moduli and effective elastic moduli of heterogeneous media. To restate this principle briefly, consider two statistically homogeneous specimens of entirely identical phase geometries. In the first the phases are all elastic with elastic moduli ${}^eC^{(r)}_{ijkl}$ of the rth phase. In the second the phases are all viscoelastic with relaxation moduli $C^{(r)}_{ijkl}(t)$ of the rth phase and consequently with TD moduli

$$\Gamma^{(r)}_{ijkl}(p) = p\hat{C}^{(r)}_{ijkl}(p). \tag{2.14}$$

Suppose that the effective elastic moduli ${}^eC^*_{ijkl}$ of the elastic specimen are known as functions of ${}^eC^{(r)}_{ijkl}$ and phase geometry. Then according to the correspondence principle the effective TD moduli Γ^*_{ijkl} are found by replacement of ${}^eC^{(r)}_{ijkl}$ by (2.14) in the known expressions for ${}^eC^*_{ijkl}$.

Let (2.11) now be introduced into (2.13) and let the result be compared with (2.5). It is easily seen that the functional dependence of B^*_{ijkl} on $i\omega$ is precisely the same as the functional dependence of Γ^*_{ijkl} on p. Therefore,

$$B^*_{ijkl}(i\omega) = \Gamma^*_{ijkl}(i\omega). \tag{2.15}$$

It should now be remembered that the p dependence enters into (2.13) through replacement of phase elastic moduli in ${}^eC^*_{ijkl}$ by (2.14). Replacement of p by $i\omega$ can simply be achieved by such a replacement in (2.14). Define

$$B^{(r)}_{ijkl}(i\omega) = \Gamma^{(r)}_{ijkl}(i\omega) = i\omega\hat{C}^{(r)}_{ijkl}(i\omega) \tag{2.16}$$

On the basis of the previous development for heterogeneous media it is seen that (2.16) are the complex moduli of the viscoelastic phases. Consequently there now emerges the following correspondence principle for effective complex moduli of viscoelastic composites: *The effective complex moduli of a viscoelastic heterogeneous specimen are found by replacement of phase elastic moduli by phase complex moduli in the expressions* ${}^eC^*_{ijkl}$ *for the effective elastic moduli of an associated heterogeneous elastic specimen, with identical phase geometry.* Symbolically

$$B^*_{ijkl}(i\omega) = {}^eC^*_{ijkl}[B^{(r)}_{mnop}(i\omega)]. \tag{2.17}$$

An incomplete version of the present correspondence principle, in the form of equation (2.15), was given by Hashin [6] and also independently by Shu [7]. Some specific results for effective complex moduli were also given in both references. Here we shall use the version (2.17) which is much more convenient.

In the event that the viscoelastic specimen has one or more elastic phases, the elastic moduli of these phases are left unchanged. It is to be noted that an elastic modulus is a degenerate case of a complex modulus, i.e. a complex modulus whose real part (elastic modulus) is frequently independent and whose imaginary part vanishes.

The analogy as given applies to anisotropic as well as to isotropic composites. In view of subsequent applications it is worthwhile to restate the analogy in some detail for isotropic composites and isotropic phases. For a statistically isotropic composite C^*_{ijkl} in (2.1) and consequently B^*_{ijkl} in (2.5) are isotropic tensors. Therefore (2.7) assumes the form

$$\tilde{\sigma}_{ij} = \tilde{\lambda}^*(i\omega)\tilde{\varepsilon}_{kk}\delta_{ij} + 2\tilde{\mu}^*(i\omega)\tilde{\varepsilon}_{ij}. \tag{2.18}$$

It is convenient to split the stresses and strains in (2.18) into isotropic and deviatoric parts. Thus

$$\tilde{\sigma}_{ij} = \tilde{\sigma}\delta_{ij} + \tilde{s}_{ij}, \qquad \tilde{\sigma} = \tfrac{1}{3}\tilde{\sigma}_{kk} \tag{2.19}$$

$$\tilde{\varepsilon}_{ij} = \tilde{\sigma}\delta_{ij} + \tilde{e}_{ij}, \qquad \tilde{\varepsilon} = \tfrac{1}{3}\tilde{\varepsilon}_{kk}. \tag{2.20}$$

Introduction of (2.19–20) into (2.18) yields

$$\tilde{\sigma} = 3\tilde{K}^*\tilde{\varepsilon} \tag{2.21}$$

$$\tilde{s}_{ij} = 2\tilde{\mu}^*\tilde{e}_{ij} \tag{2.22}$$

$$\tilde{K}^* = \tilde{\lambda}^* + \tfrac{2}{3}\tilde{\mu}^*. \tag{2.23}$$

Here $\tilde{K}^*$ and $\tilde{\mu}^*$ are the complex effective bulk and shear moduli, respectively. The relationship between different complex moduli is the same as in the theory of elasticity. Thus the effective complex Young's modulus $\tilde{E}^*$ is given by

$$\tilde{E}^* = \frac{9\tilde{K}^*\tilde{\mu}^*}{3\tilde{K}^* + \tilde{\mu}^*}. \tag{2.24}$$

Now for any isotropic viscoelastic phase there obviously hold relations such as (2.19–24). For the rth phase

$$\tilde{\sigma}^{(r)}_{ij} = \tilde{\lambda}_r(i\omega)\tilde{\varepsilon}^{(r)}_{kk}\delta_{ij} + 2\tilde{\mu}_r(i\omega)\tilde{\varepsilon}^{(r)}_{ij} \tag{2.25}$$

or

$$\tilde{\sigma}^{(r)} = 3\tilde{K}_r\tilde{\varepsilon}^{(r)} \tag{2.26}$$

$$\tilde{s}^{(r)}_{ij} = 2\tilde{\mu}_r\tilde{e}^{(r)}_{ij}. \tag{2.27}$$

The previously developed analogy now states that in order to find $\tilde{K}^*$ and $\tilde{\mu}^*$ it is necessary first to know the effective elastic moduli ${}^eK^*$ and ${}^e\mu^*$ for the associated elastic specimen. In the expressions for these moduli the elastic phase moduli ${}^eK_r$ and ${}^e\mu_r$ have then to be replaced by the complex phase moduli $\tilde{K}_r(i\omega)$ and $\tilde{\mu}_r(i\omega)$.

In view of subsequent development, different ways of writing complex moduli are recalled. Let $\tilde{M}(i\omega)$ be some complex modulus. Then

$$\tilde{M}(i\omega) = M^R(\omega) + iM^I(\omega) \tag{2.28}$$

and also

$$\tilde{M}(i\omega) = |M|\, e^{i\delta} \tag{2.29}$$

where

$$|M| = \sqrt{[(M^R)^2 + (M^I)^2]} \tag{2.30}$$

$$\tan \delta = \frac{M_I}{M_R}. \tag{2.31}$$

The angle δ in (2.31) is called the loss angle.

3. COMPLEX MODULI OF COMPOSITES

The application of the theory developed above to evaluation of complex moduli of viscoelastic composites necessitates the knowledge of expressions for effective moduli of elastic composites.

While the literature on prediction of effective elastic moduli is considerable there are available only a very small number of rigorous expressions. For discussion and review see [6]. Because of the statistical nature of the problem of prediction of effective elastic moduli, establishment of rigorous expressions is possible only for special circumstances. There are broadly two classes of special situations. The first involves some special relations between phase moduli, mostly small relative stiffness ratios, the second involves treatment of special geometry. Here the second situation is considered and in particular the geometrical model described as a composite spheres assemblage, [8]. For this model there has been derived an exact result for the effective bulk modulus K^* which may be written as

$$K^* = K_1 + \frac{(K_2 - K_1)(4\mu_1 + 3K_1)c}{4\mu_1 + 3K_2 - 3(K_2 - K_1)c}. \tag{3.1}$$

Here the subscript 2 refers to the spherical particles material, the subscript 1 refers to the matrix and c is the volume fraction of particles, [8].

According to the theory developed in Section 2 the complex bulk modulus $\tilde{K}^*(i\omega)$ of the composite spheres assemblage model is found by replacement of the phase elastic moduli in (3.1) by complex moduli. In order to keep the results simple the following assumptions are made:

(a) The particles are elastic.

(b) The matrix is viscoelastic in shear and elastic in dilatation.

These assumptions imply that K_2 and K_1 are left unchanged in (3.1) while μ_1 is to be replaced by

$$\tilde{\mu}_1(i\omega) = \mu_1^R(\omega) + i\mu_1^I(\omega) = \mu_1^R(\omega)[1 + i \tan \delta_\mu^{(1)}] \tag{3.2}$$

Here the matrix shear loss angle $\delta_\mu^{(1)}$ is defined by

$$\tan \delta_\mu^{(1)} = \frac{\mu_1^I}{\mu_1^R}. \tag{3.3}$$

It shall furthermore be assumed that

$$\tan^2 \delta_\mu^{(1)} \ll 1. \tag{3.4}$$

This is justified for many materials, such as polymers below the glass transition temperature in which the loss tangent is generally below 0·1. There is, of course, no difficulty to

carry out the following analyses without the assumption (3.4). This merely results in more cumbersome results.

Introducing (3.2) into (3.1), separating into real and imaginary parts and using (3.4), the following results are obtained

$$K^{*R} = K_1 + \frac{(K_2 - K_1)(4\mu_1^R + 3K_1)c}{4\mu_1^R + 3K_2 - 3(K_2 - K_1)c} \tag{3.5}$$

$$K^{*I} = \left[\frac{K_2 - K_1}{4\mu_1^R + 3K_2 - 3(K_2 - K_1)c}\right]^2 12\mu_1^I c(1-c) \tag{3.6}$$

where

$$K^*(i\omega) = K^{*I}(\omega) + iK^{*R}(\omega).$$

It is seen that K^{*R} as given by (3.5) is the elastic bulk modulus (3.1), with μ_1 replaced by μ_1^R. Since μ_1^R is in general frequency dependent, so is K^{*R}.

Expression (3.6) shows that K^{*I} vanishes for $c = 0, 1$ as it should. For when $c = 0$ there are no inclusions which leaves a homogeneous viscoelastic matrix, which by hypothesis shows no viscoelastic effects in dilatation. When $c = 1$ there is no matrix and hence no viscoelastic effect.

It is also interesting to note that the presence of the elastic inclusions in a viscoelastic matrix, which is elastic in dilatation and viscoelastic in shear introduces a macroscopic dilatational viscoelastic effect. A similar result has been found for analysis of static viscoelastic dilatational behavior of the same composite model, [1]. This phenomenon is to be expected for any phase geometry.

Let it now be assumed that the inclusions are perfectly rigid. This is a good approximation for materials such as reinforced solid propellants in which the reinforcing particles are by an order of magnitude stiffer than the matrix. In that event

$$\mu_1^R \quad \text{and} \quad K_1 \ll K_2 \tag{3.7}$$

and equations (3.5–6) become

$$K^{*R} = \frac{1}{1-c}\left(K_1 + \frac{4}{3}\mu_1^R c\right) \tag{3.8}$$

$$K^{*I} = \frac{c}{1-c}\frac{4}{3}\mu_1^I \tag{3.9}$$

Therefore

$$\tan \delta_K^* = \frac{K^{*I}}{K^{*R}} = \frac{4\mu_1^I c}{3K_1 + 4\mu_1^R c}. \tag{3.10}$$

Examination of (3.10) reveals a difficulty when c approaches unity. In that case there is no matrix left and the effective loss tangent must vanish, but it is seen that for $c = 1$, (3.10) retains a finite value which still depends on non-existent matrix properties. The reason for the difficulty is that the limiting process leading from (3.6) to (3.10) by use of (3.8) becomes invalid when $c \approx 1$. The conclusion is that for very stiff particles and very high particle volume fraction K^{*I} should be computed by use of (3.6) with the actual values of μ_1^R, K_1 and K_2, and the rigid particle approximation is not permissible.

Another interesting special case is a porous material. In that case

$$K_2 = 0 \tag{3.11}$$

and it follows from (3.5–6) and (3.11)

$$K^{*R} = \frac{4K_1\mu_1^R(1-c)}{4\mu_1^R + 3K_1 c} \tag{3.12}$$

$$K^{*I} = \left[\frac{K_1}{4\mu_1^R + 3K_1 c}\right]^2 12\mu_1^I c(1-c) \tag{3.13}$$

$$\tan\delta_K^* = \frac{K^{*I}}{K^{*R}} = \frac{3K_1 c}{4\mu_1^R + 3K_1 c}\tan\delta_\mu^{(1)}. \tag{3.14}$$

We now turn to discussion of the complex shear modulus and we first derive some interesting general results for two special situations. One is a viscoelastic incompressible matrix which is reinforced by perfectly rigid particles, the other is a viscoelastic incompressible porous material. In the associated elastic cases, with same phase geometry the effective shear modulus must have the form

$$\mu^* = \mu^*(\mu, \text{phase geometry}), \tag{3.15}$$

where μ is the matrix shear modulus. By dimensional argument equation (3.15) can be put into the following form

$$\mu_r^* = \mu\psi_r \tag{3.16}$$

$$\mu_p^* = \mu\psi_p \tag{3.17}$$

where ψ is a nondimensional function of phase geometry only and the subscripts r and p indicate different functions for rigid reinforcement and porous material, respectively. By the correspondence principle for complex moduli as developed in Section 2 it follows immediately that for the viscoelastic specimens

$$\mu_r^*(i\omega) = \mu(i\omega)\psi_r \tag{3.18}$$

$$\mu_p^*(i\omega) = \mu(i\omega)\psi_p. \tag{3.19}$$

Separation of (3.18–19) into real and imaginary parts yields

$$\mu_r^{*R} = \mu^R\mu_r, \qquad \mu_r^{*I} = \mu^I\psi_r \tag{3.20}$$

$$\tan\delta_{\mu_r}^* = \tan\delta_\mu \tag{3.21}$$

$$\mu_p^{*R} = \mu^R\psi_p, \qquad \mu_p^{*I} = \mu^I\psi_p \tag{3.22}$$

$$\tan\delta_{\mu_p}^* = \tan\delta_\mu. \tag{3.23}$$

These results show that the loss tangents of the nonhomogeneous materials are the same as the matrix loss tangents in the cases considered and that both real and imaginary parts of the complex effective shear modulus are related to their matrix counterparts precisely as an effective elastic shear modulus to matrix shear modulus.

For an incompressible matrix the complex Young's modulus $E(i\omega)$ is related to the complex shear modulus by

$$E^R(\omega) = 3\mu^R(\omega), \qquad E^I(\omega) = 3\mu^I(\omega). \tag{3.24}$$

For rigid reinforcement the composite is also incompressible. Therefore the effective complex Young's modulus and shear modulus of the composite also obey a relationship as (3.24). It therefore follows that

$$E_r^{*R} = E^R\psi_r, \qquad E_r^{*I} = E^I\psi_r \tag{3.25}$$

$$\tan\delta_{E_r}^* = \tan\delta_E = \tan\delta_\mu \tag{3.26}$$

where ψ_r in (3.25–26) is the same function which appears in (3.16). It is not clear that relations of type (3.25–26) can be obtained also for a porous material for such a material is not necessarily macroscopically incompressible.

The special cases considered above do not lead to appreciable simplification when it is desired to obtain results for μ^{*R} and μ^{*I}. We therefore consider the general case of a two phase composite. In order to exploit the correspondence principle it is necessary to have rigorous results for μ^* of an elastic two phase material for some geometry. Simple closed form results for the effective shear modulus have been derived for dilute suspensions of spherical particles [8, 9]. There is no difficulty to use the present correspondence principle to derive similar results for the complex shear modulus of dilute spheres suspensions. An example for such a calculation may be found in [10]. Unfortunately, however, dilute suspensions are chiefly of academic interest and therefore this subject will not be pursued here.

We consider again the composite spheres assemblage model. In the original treatment of this model in [8] only upper and lower bounds for the effective shear modulus were derived. It has recently been shown by Christensen [11] that these bounds can be exploited to construct similar upper and lower bounds for the effective complex shear modulus for this model.

Recent work by Hashin and Rosen (to be published) indicates that the upper bound for μ^* derived in [8] may actually be the correct solution for the effective shear modulus, for particles which are stiffer than the matrix. It has also been shown that the upper bound of [8] can be brought into the explicit form

$$\mu^* = \mu_1\left[1+\frac{c}{\dfrac{1}{\gamma-1}+A(1-c)-\dfrac{c(1-c^{\frac{2}{3}})^2}{Bc^{\frac{2}{3}}+C}}\right] \tag{3.27}$$

where

$$\gamma = \frac{\mu_2}{\mu_1} \tag{a}$$

$$A = \frac{2(4-5\nu_1)}{15(1-\nu_1)} \tag{b}$$

$$B = \frac{10}{21}(1-\nu_1)\frac{(7-10\nu_2)(7+5\nu_1)-\gamma(7-10\nu_1)(7+5\nu_2)}{4(7-10\nu_2)+\gamma(7+5\nu_2)} \tag{c}$$

$$C = \frac{10}{21}(7-10\nu_1)(1-\nu_1). \tag{d}$$

(3.28)

Here ν_1 and ν_2 are the Poisson's ratios of matrix and particles, respectively, and c is the volume fraction of the spherical particles.

Now suppose that the particles are elastic and that the matrix is viscoelastic. Then, according to the correspondence principle the matrix elastic constants have to be replaced by their complex counterparts. In this respect it is recalled that

$$\nu = \frac{3K - 2\mu}{2(3K + \mu)}. \tag{3.29}$$

If it is further assumed for simplicity that the matrix is viscoelastic only in shear we have from (3.29), for the matrix

$$\nu_1(i\omega) = \frac{3K_1 - 2\tilde{\mu}_1(i\omega)}{2[3K_1 + \tilde{\mu}_1(i\omega)]}. \tag{3.30}$$

Thus, in order to obtain $\tilde{\mu}^*(i\omega)$ it is necessary to replace μ_1 and ν_1 in (3.27–28) by $\tilde{\mu}_1(i\omega)$ and (3.30), respectively. It is unfortunately also seen that because of the complexity of (3.27–28), subsequent separation into real and imaginary parts requires very heavy algebra.

Consider the special but very appropriate case (3.4). In this case it is possible to prove some interesting general results. Let the effective elastic shear modulus of *any* two phase elastic material be denoted ${}^e\mu^*(K_1, \mu_1, K_2, \mu_2, \text{geometry})$. Let μ_1 be replaced by μ_1^R and let the resulting expression be denoted

$${}^e\mu^*(K_1, \mu_1^R, K_2, \mu_2, \text{geometry}) = {}^e\mu^*(\mu_1^R). \tag{3.31}$$

Let $\tilde{\mu}^*(i\omega)$ be written in the usual form

$$\tilde{\mu}^*(i\omega) = \mu^{*R}(\omega) + i\mu^{*I}(\omega) \tag{3.32}$$

and

$$\tan \delta^* = \frac{\mu^{*I}}{\mu^{*R}}. \tag{3.33}$$

It may then be shown that if (3.4) is fulfilled

$$\mu^{*R} = {}^e\mu^*(\mu_1^R) \tag{3.34}$$

$$\tan \delta^* = \frac{\mu_1^R}{{}^e\mu^*} \frac{\partial {}^e\mu^*}{\partial \mu_1^R} \tan \delta_\mu^{(1)}. \tag{3.35}$$

Details of the derivation will be published elsewhere.

The meaning of (3.34) is that the real part of the effective shear modulus is simply found by replacement of the matrix shear modulus by the (frequency dependent) real part of the matrix complex shear modulus. Therefore, for the composite spheres assemblage model $\mu^{*R}(\omega)$ is found by replacement of μ_1 by $\mu_1^R(\omega)$ in (3.27–28). It is noted in this respect that because of (3.30) and (3.4), ν_1 in (3.27–28) is now to be interpreted as

$$\nu_1(\omega) = \frac{3K_1 - 2\mu_1^R(\omega)}{2[3K_1 + \mu_1^R(\omega)]}.$$

The use of (3.35) is unfortunately not as simple and calls for a tedious differentiation of (3.27) which will not be performed here.

It is mentioned in passing that formulae similar to (3.34) apply for the effective complex bulk modulus of a two phase material in which one phase is elastic, the other is viscoelastic in shear only and (3.4) is fulfilled. As a check it is easily verified that (3.5–6) can be obtained from (3.1) by use of such formulae.

Some experimental verification of the present results is provided by experiments carried out by Bodner and Lifshitz [12] who measured dynamic viscoelastic properties of a composite consisting of silica sand particles imbedded in an epoxy matrix. Pertinent material data are given below:

Silica sand, (2)

$$v_2 = 0{\cdot}38$$

$$\mu_2 = 4{\cdot}14 \times 10^6 \text{ psi} \qquad \nu_2 = 0{\cdot}25.$$

Epoxy, (1)

$$v_1 = 0{\cdot}62$$

$$\mu_1^R = 1{\cdot}12 \times 10^5 \text{ psi} \qquad \text{at frequency 18 cps}$$

$$\nu_1 = 0{\cdot}42 \qquad \text{quasi-static value.}$$

The real part of the epoxy complex Young's modulus varied linearly with log. frequency. Over a range of 10 to 10^5 cps the real part increased only by about 50%.

It was found experimentally that

$$\frac{E^{*R}(\omega)}{E_1^R(\omega)} = 3{\cdot}0 \tag{3.36}$$

with high accuracy over the whole frequency range. It was furthermore found that

$$\tan \delta_E^* \sim \tan \delta_E^{(1)} \tag{3.37}$$

over the entire frequency range, i.e. the composites' extensional loss tangent was approximately equal to the matrix loss tangent.

It is seen that (3.37) and the frequency independence of the ratio in (3.35) agree with the conclusions (3.25–26), in spite of the fact that the ideal conditions on which (3.25–26) are based were not fulfilled in the experiment.

In order to take better account of the material properties of the sand–epoxy composite, μ^{*R} has been computed from (3.27–28), (3.34) and K^{*R} from (3.6). Since the composites' loss tangents are also small it follows from (2.24) that

$$E^{*R} \cong \frac{9K^{*R}\mu^{*R}}{3K^{*R}+\mu^{*R}}$$

which is the usual elastic formula in terms of real parts.

Computation of the ratio (3.36) for the material properties of the phases listed above gave

$$\frac{E^{*R}(\omega)}{E_1^R(\omega)} = \begin{cases} 3{\cdot}0 & \text{for} \quad 18 \text{ cps} \\ 2{\cdot}8 & \text{for} \quad 100{,}000 \text{ cps} \end{cases}$$

the ratio being monotonically decreasing with frequency. This is in excellent agreement with the experimental result (3.36).

The loss tangent $\tan \delta_E^*$ has not been computed in detail but an order of magnitude estimation based on (3.35), (3.27–28) and (3.5–6) shows that it is very nearly equal to the matrix extensional loss tangent. This is in agreement with (3.37).

It should be carefully noted that assumption of rigid particles and/or incompressible matrix in (3.27–28) may lead to serious errors in the prediction of the value of μ^{*R}. The simplification of (3.27–28) in these special cases is

Rigid particles

$$\mu^* = \mu_1 \left[1 + \frac{\dfrac{15(1-\nu_1)}{2(4-5\nu_1)} c}{1 - c\left[1 + \dfrac{63}{4(7-10\nu_1)(4-5\nu_1)} \phi(c)\right]} \right] \quad \text{(a)}$$

$$\phi(c) = \frac{(1-c^{\frac{2}{3}})}{1-c^{\frac{7}{3}}}. \quad \text{(b)} \qquad (3.38)$$

Rigid particles—incompressible matrix

$$\mu^* = \mu_1 \left[1 + \frac{2{\cdot}5c}{1 - c\left[1 + \frac{21}{4}\phi(c)\right]} \right]. \qquad (3.39)$$

For comparison the numerical values of μ^{*R}/μ_1^R predicted by the different formulae are given below for material properties at 18 cps.

Equation	μ^{*R}/μ_1^R
(3.27–28)	3·16
(3·37)	3·60
(3·38)	9·10

It is seen that the incompressible matrix assumption leads to a very serious error.

Various approximate expressions for the effective shear modulus of elastic two phase materials have been proposed in the literature. Invariably, these are based on simplifying assumptions whose validity is difficult to assess. It is appropriate to mention here a result given by Van der Poel [13] since both Bodner and Lifshitz [12] and Schwarzl *et al.* [14] have reported good agreement between their experimental results and the Van der Poel prediction.

The method used in [13] is an example of an approximation which has been called "self consistent scheme" by various writers. It may be shown that an effective shear modulus can be determined if the average shear strain in one phase of a sheared composite is known. In the [13] version the shear strains in the particles are estimated by consideration of a typical spherical particle as if it were imbedded in a concentric spherical matrix shell. The resulting composite sphere is then thought to be imbedded in an infinite equivalent homogeneous material whose elastic moduli are assumed to be the effective moduli of the composite. This defines a three layer boundary value problem to determine the strain in the particle and two separate calculations are needed to determine the effective bulk and shear modulus.

The effective bulk modulus predicted in [13] is curiously the same as the result (3.1). The effective shear modulus calculation is, however, incorrect since the solution of the boundary value problem mentioned above (equations (6–11) in Ref. [13]) does not satisfy

the equations of elasticity. Also comparison with spherical shell elasticity solutions, as given for example in Lur'e [15], shows that irrational r powers as enter into Van der Poel's solution are incorrect.

With respect to the reported agreement between experiments and the Van der Poel result it should be noted that the theoretical results were taken from a table in [13] which was computed for incompressible matrix. Now the matrix in the [12] experiments was not incompressible ($\nu_1 = 0{\cdot}42$) and it has been shown above that the incompressibility approximation is not permissible for the prediction (3.27–28). It would seem therefore that the Van der Poel result should be evaluated for the proper matrix Poisson's ratio for purpose of comparison with experiments. It is, however, not clear to the writer whether this is a worthwhile undertaking since the necessary numerical calculations are very heavy and the underlying theory is clearly incorrect.

4. CONCLUSION

Effective complex moduli of viscoelastic composites have been defined within the frame of a first order approximation to the dynamic macro-behavior of viscoelastic composites. It has been shown that such effective complex moduli are related by a simple correspondence principle to effective elastic moduli of composites.

Results for complex moduli have been derived for the composite spheres assemblage model and for general incompressible–rigid and incompressible–porous viscoelastic composites. Good agreement between present theoretical results and experiments described in [12] has been obtained.

In an accompanying paper a similar approach is used for prediction of complex moduli of viscoelastic fiber reinforced materials.

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Абстракт—Выводится соответствующий принцип позваляющий определить эффективные комплексные модули вязкоупругих смесей, на основе аналитических выражений для эффективных упругих модулей смесей. Метод применяется для определения комплексных модулей изотропных многокорпускулярных смесей, со специальным учетом для сборной модели смешанных шариков. Получается очень хорошая сходимость с публикованными экспериментальными результатами для эффективных комплексных модулей.